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A unique facility for surface microscopy

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Abstract

The Advanced Surface Microscopy Facility at the University of California at Davis is a unique collection of ultrahigh vacuum surface microscopy and analysis instruments, including a low energy electron microscope, a variable-temperature scanning tunneling microscope, and an X-ray photoelectron spectrometer. The system design has features for optimal sample transfer and handling among the primary instruments. The performance of the system is shown through LEEM images of clean Mo(100) and STM images of rutile TiO_2.

Keywords: Surface microscopy; Ultrahigh vacuum; STM; LEEM

1. Instrumentation components

The primary ultrahigh vacuum (UHV) surface microscopy and analysis instruments which comprise the Advanced Surface Microscopy Facility (ASMF) at the University of California at Davis are as follows: an Elmitec low energy electron microscope (LEEM), an Oxford variable-temperature scanning tunneling microscope (VT-STM), and a Vacuum Generators X-ray photoelectron spectrometer (Fig. 1). Each of these instruments is installed in a separate UHV chamber, with the three main chambers and three minor ones connected together and separated by gate valves. The system design permits the motion of samples throughout the interconnected UHV system. The sample holder for the Oxford STM was specially designed to be compatible with that of the Elmitec LEEM. One airlock is attached to the LEEM and allows the entrance of samples to a small preparation chamber. A long magnetic transfer bar (motion ~ 1.1 m), which has a bayonet mechanism for engaging the sample, moves the sample from the preparation chamber into the LEEM or onto the manipulator in the XPS chamber. A second airlock allows the entrance and exit of samples and the two interchangeable scanner/tip assemblies directly into the Oxford STM. A second long magnetic transfer bar (motion ~ 0.94 m) can move the sample from the STM, to the manipulator in the XPS chamber, or onto a sample parking place assembly located in the pumping elbow adjacent to the STM.

1.1. Low energy electron microscope (LEEM)

The primary instrument in the ASMF is the LEEM, a type of instrument designed originally by Bauer [1,2]. This LEEM instrument was designed and built by Elmitec Elektronenmikroskopie in Clausthal-Zellerfeld, Germany. A photograph of the LEEM and XPS chambers is shown in Fig. 2. The LEEM is capable of real-time, real-space imaging of metals and semiconductors, during sample heating or cooling and deposition of adsorbates. This instrument comes with a special sample holder (Fig. 3), which allows the sample to be heated as hot as 2900 K. The imaging stage can also be cooled with liquid nitrogen to achieve sample temperatures as low as 120 K. Contrast in images produced by this instrument arises from many sources, including differing surface structures, atomic species, and surface geometry. This instrument can also be used to obtain low energy electron diffraction (LEED) patterns, which are used to analyze surface structures. At zero sample voltage, mirror electron microscopy (MEM) images can be

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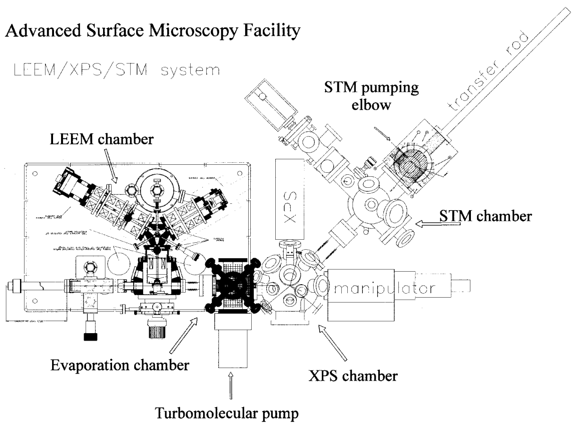

Fig. 1. Plan view of schematic drawing of the ASMF, including the LEEM, XPS, STM, and attendant chambers. (LEEM chamber schematic drawing courtesy of Elmitec Elektronenmikroskopie; STM chamber schematic drawing courtesy of Oxford Instruments.)

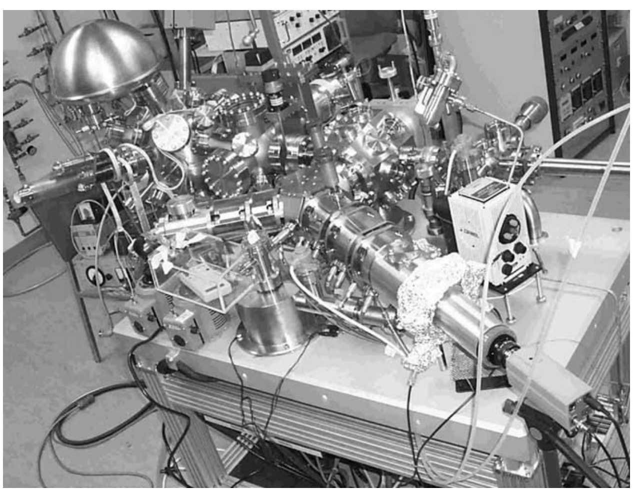

Fig. 2. Photograph of LEEM and XPS chambers, as viewed from the rear. The LEEM layout is evident. Note that the photograph was taken before the installation of the STM.

measured. Using an ultraviolet mercury lamp as a source of photons, photoelectron emission microscopy (PEEM) images can also be measured in this instrument on all samples, regardless of surface ordering.

1.2. Variable temperature scanning tunneling microscope (VT-STM)

The scanning tunneling microscope, or STM, is capable of atomic resolution, real-space imaging of metal and semiconductor surfaces [3]. The VT-STM instrument of the ASMF was constructed by Oxford Instruments in Cambridge, England and was designed to image within a sample temperature range of 80–1000 K (Fig. 4). The STM's capabilities nicely complement the LEEM's, as it can achieve atomic resolution and can image structures on smaller scales than those detectable by LEEM or LEED. In addition, STM can be used to elucidate additional information, such as adsorbate binding sites and molecular orientations. In contrast to the LEEM, however, STM images typically take seconds to minutes to achieve, and thus are much slower in time scale than those from the video-rate LEEM.

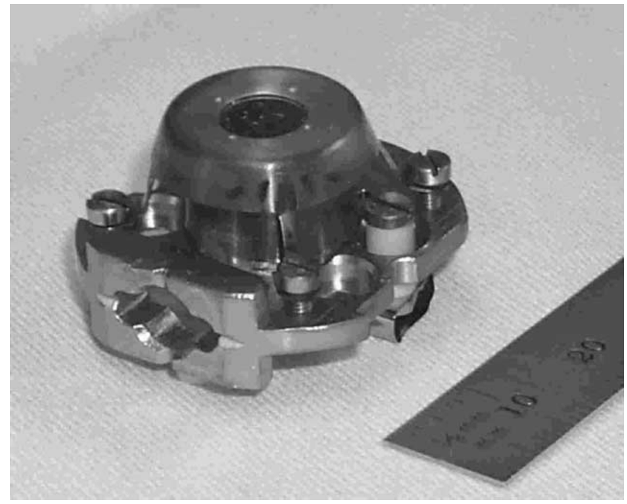

Fig. 3. Photograph of LEEM sample holder, with millimeter ruler in the foreground to show the lateral scale. The photograph shows the molybdenum cap over a Cu(111) sample. The baseplate of the sample holder is made from titanium. A W-25% Re/W-5% Re thermocouple is attached to a Mo ring directly beneath the sample. Further below the sample is a tungsten electron beam filament, surrounded by a Mo reflector cup.

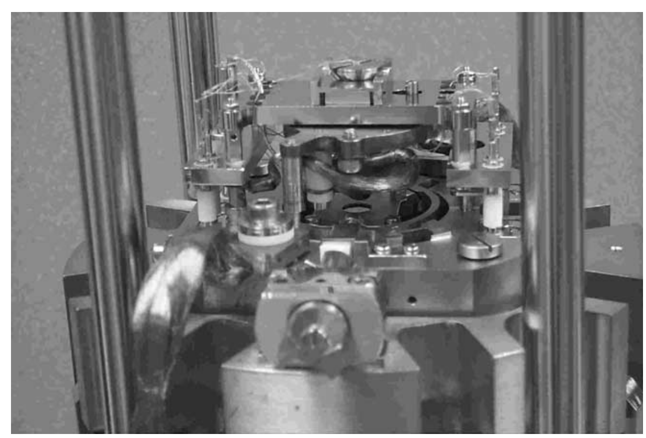

Fig. 4. Photograph of Oxford Instruments VT-STM. Sample surface is in center, with screw to locate position of sample surface in foreground. Scanner assembly is on top, but the piezoelectric scanner is hidden by the conical radiation shield. Vertical tubular supports contain springs for vibration isolation of the sample stage from the supporting flange.

1.3. X-ray photoelectron spectrometer (XPS) and sample cleaning and preparation equipment

To round out the surface analysis capabilities of the ASMF, a Vacuum Generators X-ray photoelectron spectrometer (XPS) is located in the surface analysis chamber between the STM and LEEM chambers. This VG Microtech XPS system consists of an XR3E2 dual anode Mg/Al X-ray source, a VG 100AX electron energy analyzer, and accompanying electronics and software. The XPS is used for identification of chemical species on surfaces. Since neither of the other instruments is capable of direct chemical identification, XPS enables the quantitative study of the effects of varied concentration on various phases of adsorbed species, as well as providing information about chemical bonding. The XPS surface analysis chamber also includes a sample manipulator for heating, cooling, and positioning the sample, an argon ion sputtering gun (OCI IONEC Model IG35) for cleaning the sample, and a mass spectrometer (Stanford Research Systems RGA 200) for residual gas analysis from 1 to 200 amu. The chamber also has ports for optical signals to enter and leave so that laser spectroscopy, such as sum frequency generation, could be used as a method of measuring vibrational spectra on the surfaces.

2. System design features

2.1. Vibration isolation

For surface microscopies, isolation of the microscope from building and other vibrations is important to maximize resolution and decrease noise. Both the LEEM and STM instruments require vibration isolation in order to achieve optimum resolution. The LEEM was delivered with spring-supports within the table on which it sits, which is sufficient isolation for it. The STM system as delivered, however, did not have any vibration isolation outside of the vacuum chamber, although some vibration dampening measures were designed into the microscope stage within the chamber. Since it would be only partially isolated from the XPS chamber by the welded bellows connecting the two, the STM and XPS chambers were placed together on a steel framework designed by Anderson [4]. This framework was supported by three air-filled vibration isolation legs (I-2000 Stabilizer™ Pneumatic Vibration Isolator, Newport Corporation, Irvine, CA). The same framework also supports the sample manipulator on the XPS chamber and the transfer bar on the STM chamber. To ensure that the chambers supported by the framework and the LEEM chamber could retain a measure of vibrational independence, the evaporation chamber and the LEEM chamber were connected using a length of edge-welded bellows in order to vibrationally decouple the chambers.

2.2. Sample transfer

The special LEEM sample holder, as delivered by Elmitec, enables not just sample manipulation but also sample heating. Although the base plate is made of titanium, most other sample holder components are made of molybdenum, both to withstand high sample temperatures and to insure that the sample holder remains non-magnetic. Heating is accomplished by a tungsten filament within the sample holder, which is electrically isolated from the rest of the sample holder. This enables heating either by radiation from the hot filament or by electron bombardment with a voltage bias between the sample and the filament. A molybdenum reflecting cup, beneath and around the filament, held at the same voltage as the filament, directs all of the radiation or electrons at the sample. This sample holder also has features which uniquely enable LEEM imaging, such as a highly polished, smooth molybdenum cap, which is designed to decrease field-emission and maintain electric field homogeneity near the sample during imaging. It also contains a tungsten–25% rhenium/tungsten–5% rhenium thermocouple attached to a small molybdenum ring which supports the sample, allowing for sample temperature monitoring. Small loops of tungsten–rhenium foil form the electrical contacts for the sample (two for the filament current and two for the thermocouple), making contact with pins which protrude from the sample stage.

As the sample holder needs to transport the sample into each of the three major instruments in the ASMF, sample stages which could accommodate this unique sample holder were designed. For the STM, Oxford

Fig. 5. Photograph of Oxford Instruments top plate for sample holder. The modified top plate has a screw to locate the position of the sample surface with respect to the rest of the STM so as to minimize sample drift as the temperature is varied.

Instruments redesigned both the sample stage and the sample holder top plate. Oxford's STM design requires a thick molybdenum top plate (see Fig. 5) so that a screw can register the position of the top surface of the sample with respect to the tip assembly, reducing thermal drift in the instrument. Oxford's instrument design also involves using a wobblestick to grab the thicker top plate of the sample holder using pincers, in order to drop it into the sample stage. The surface of this top plate can still be well-polished to decrease field emission from the top plate during LEEM imaging, where there is a high electric field between the top plate (at 20 kV) and the objective lens (at 0 V). In addition, the shape was designed so as to have no surfaces or sharp edges within 2 mm of the objective lens, in order to prevent arcing of the top plate to the objective lens during LEEM imaging.

Oxford also redesigned their original sample stage to accommodate the electrical contacts on the LEEM sample holder. They included spring-loaded copper contacts which provide electrical contact and also press the sample holder up against reference points, to help preserve the excellent drift specification of this instrument.

A number of additions to the XPS and STM chambers were also necessary to make sample transfer possible. Because of the design of the XPS chamber, ports with axes at 135° were chosen for transfer into and out of the chamber (see Fig. 1). This required rotation of the sample holder by this angle, since the LEEM sample holder can be engaged by a transfer bar on one side only. This rotation was accomplished by designing an azimuthal sample rotation stage into the sample manipulator. This manipulator is also capable of polar rotation (about its long (z) axis), and has motion of 4 inches in the z direction (into and out of the chamber) and ± 0.5 inches in each of the x and y (horizontal and vertical) directions. Its design is shown schematically in Fig. 6(a) and (b), with a photograph in Fig. 6(c). The gears are driven by a rod connected to a rotary feedthrough. The manipulator is also equipped with a dewar which can be filled with liquid nitrogen. This dewar is connected by copper braids to the sample stage, where the sample can be cooled through the sample holder. The entire manipulator is supported by a long tube connected to an XYZ stage which allows the manipulator to be moved in the chamber, and a rotary stage, which allows polar rotation (along the long axis of the support tube, which passes through the baseplate of the LEEM sample holder when mounted). This great amount of flexibility in motion and position allows the user to compensate for slight misalignments of the chamber transfer axes, perform angle-resolved XPS, and change the position of the sample to put it in the optimum position in front of the various instruments (sputter gun, XPS, mass spectrometer) installed on that

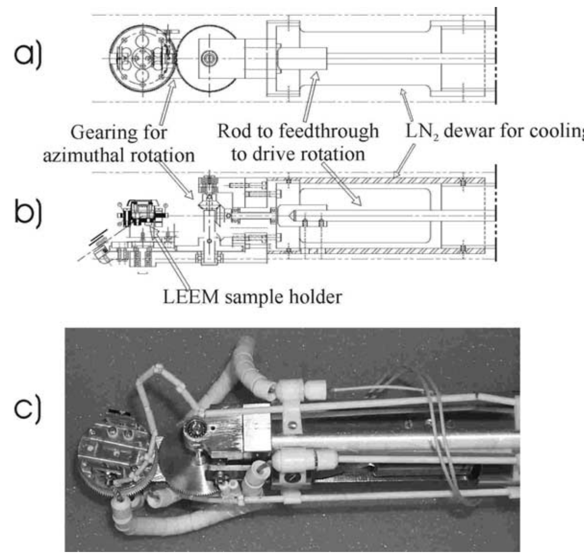

Fig. 6. Schematic drawings and photograph of the end of the sample manipulator in the XPS chamber. (a) Top view of schematic drawing. (b) Side view of schematic drawing. (c) Photograph of end of sample manipulator, viewed from top. The drawings show the gearing which enables the azimuthal rotation, as described in the text. The original LEEM sample holder is shown in place in (b). The mechanism for sample rotation and cooling was designed by T. Anderson, UC Davis Department of Physics. (Sample holder drawing courtesy of Elmitec Elektronenmikroskopie GmbH.)

chamber, as well as positioning the sample for viewing from the outside. The sample stage on this manipulator is also equipped with electrical contacts, as in the LEEM and STM chambers, to enable sample heating and temperature monitoring.

Another addition to the XPS chamber was required because of the long distance between the XPS and LEEM chamber, necessitated by the width of the table on which the LEEM chamber is installed (see Fig. 1). A length of welded bellows was installed to decrease the transmission of vibration between the LEEM and XPS chambers, which is necessary as they have separate support structures. Additional space is occupied by an "evaporation chamber," which provides ports for attachment of a turbomolecular pump, with gate valve and large tube diameters along the path to the XPS chamber, to ensure good conductance of gases. This chamber also has ports designed for metal evaporators and leak valves. In addition, another sample stage could be installed on this chamber for more convenient gas dosing or evaporation.

Because the STM chamber lacked a downward-pointing flange, a separate chamber was designed and coupled adjacent to the STM chamber. Onto this "pumping elbow" were placed additional ports for an ionization gauge, titanium sublimation pump, leak valves, windows, and the transfer bar for moving samples from the XPS chamber. Another port was

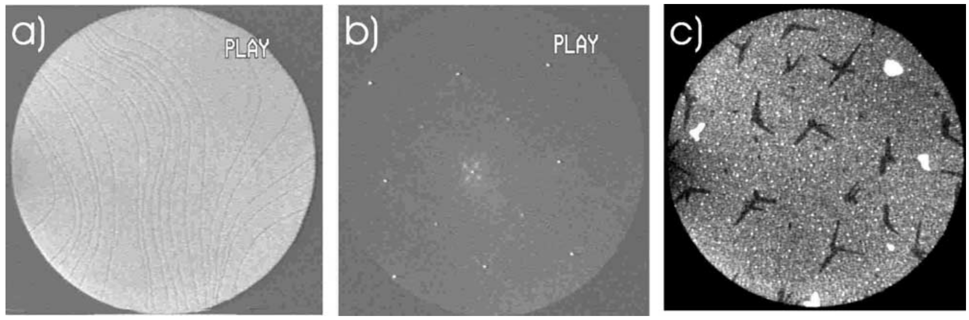

Fig. 7. (a) LEEM image of clean Mo(100), showing very clear atomic steps and demonstrating resolution of 10 nm. Field of view is 2 μm, and the imaging electron energy is 5 V. (b) LEED image of Mo(100) measured at 65 V in the LEEM, showing the expected square pattern, with some extra superstructure spots in the center corresponding to the presence of carbon islands. (c) Mo(100) image measured during the deposition of Pb, with 30 μm field of view and 5 V imaging electron energy. The small bright spots correspond to the Pb, and the large dark features are carbon islands.

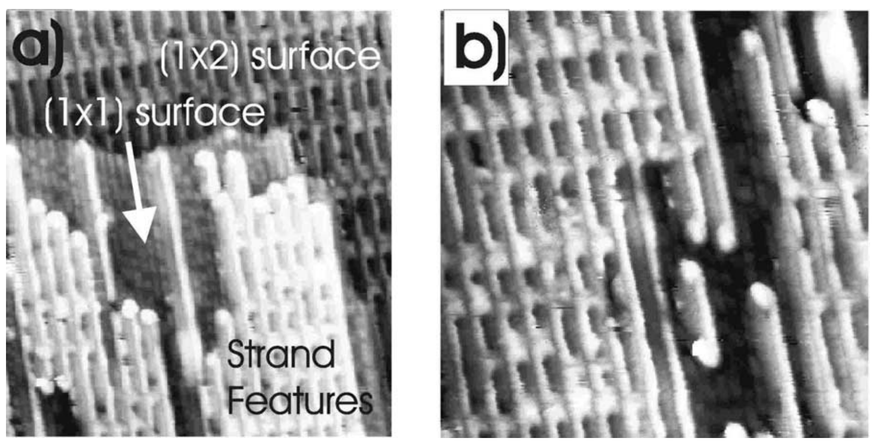

Fig. 8. STM images of rutile $TiO_2(110)$. (a) 250 × 290 Å. Regions of (1 × 1) and (1 × 2) surface structure, as well as strand features, are indicated. (b) 200 × 200 Å. Sample bias is +1.7 V and tunneling current is 0.18 nA for both images.

added for a sample ‘‘parking place,’’ which is simply two vertical plates with slots to hold up to four sample holders, mounted on a ‘‘push-pull’’ linear motion device. This is situated in a vertical port above the sample transfer axis, so that the parking place can be lowered through the transfer bar axis and have samples placed in it. The parking place can then be retracted to store the samples above the transfer bar axis.

3. System performance

3.1. LEEM images of clean Mo(100)

Clean Mo(100) is a good prototypical system for demonstrating the properties of the LEEM. It is easy to clean, requiring only a high temperature flash to 2000 K. Fig. 7(a) shows a LEEM image of Mo(100) which clearly resolves atomic steps on the surface. The nominal resolution of this LEEM instrument is 10 nm. Fig. 7(b) shows a LEED image measured on the same surface. The important ability of the LEEM to be able to image the sample during the growth of an overlayer is demonstrated in Fig. 7(c), which shows Pb (bright white spots) on Mo(100) during the deposition process.

3.2. STM images of clean rutile TiO_2 (110)

The inertness of many metal oxide surfaces to chemical reactions makes them ideal as supports in heterogeneous catalysis. Nevertheless, such surfaces exhibit some degree of reactivity and it is widely believed that this is a result of defects. Rutile $TiO_2(110)$ has emerged as a prototypical metal oxide surface, and several studies have been undertaken to explore its surface structure [5–7]. Several types of reconstructions and surface defects have been observed with STM, including a (1 × 2) phase and thick strand features oriented along the distinctive [001] crystal direction (Fig. 8).

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