Integrated cryogenic scanning tunneling microscopy and sample preparation system

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A scanning tunneling microscope (STM) operable from room temperature to 1.5 K with an integrated sample preparation chamber has been constructed and successfully tested. Both the sample and the tunneling tip can be cleaned and/or modified in a UHV processing chamber which is connected at the top of a liquid-helium flow cryostat. The STM is mounted to a threaded rod inside the cryostat so that it can be translated vertically. This allows a newly prepared sample/tip to be transferred onto the STM and lowered into a cryogenic environment without breaking vacuum, minimizing surface contamination which causes significant tunneling problems at cryogenic temperatures.

I. INTRODUCTION

The development of the scanning tunneling microscope (STM) with its capability to resolve both topology and electronic structure on the atomic scale represents a major advance in surface science. The STM has been applied to a large variety of experiments in the fields of physics, chemistry, engineering, and biology, many of which require successful operation at low temperatures.

Exposing a sample surface to laboratory air inevitably results in the absorption of some contaminants. In performing STM at room temperature, minimizing contamination is essential as it obscures the desired surface and can damage the tunneling tip. At cryogenic temperatures we have found that surface cleanliness becomes even more of a challenge: not only are contaminants more likely to stick to the surface, their presence can cause an even greater degradation in STM performance.

Depending on the amount of exposure to air, metal surfaces may absorb one or more monolayers of contaminants (e.g., oxides, water) which are largely transparent to the STM at room temperature. Although it seems improbable that the STM would work at all with the tunneling tip poking through this layer of dirt, indirect evidence suggests that this commonly occurs. As long as the layer is insulating and the tunneling tip can move through it without being altered, there is little degradation of the STM performance. However, upon cooling, this layer freezes solid and severely affects the delicate tunneling tip. This causes extremely unstable behavior when performing cryogenic STM even though atomic resolution images can often be obtained on the same sample at room temperature. Clearly, it would be advantageous to develop a system which allowed a newly prepared surface to be probed without ever being exposed to air.

II. DESIGN

Our goal was to build an STM system featuring in situ cleaning and modification of the sample and the tip, which would be suitable for the study of low and high temperature superconductor crystals, films, and microfabricated devices. In order to perform topographic and spectroscopic measurements near the relevant transition temperatures and to achieve an energy resolution ~100 µV, the system would need to be operable from room temperature down to ~1 K. Also, in order to study the effects of a high applied magnetic field, the STM would need to be situated in the bore of a superconducting magnet. To satisfy these criteria, we built a system featuring a compact STM mounted in a liquid helium cryostat with an adjacent processing chamber in which the tip and sample can be prepared, as shown in Fig. 1(a). The STM features a horizontal tip and a vertical sample, both of which can be transferred between the microscope and the processing chamber.

The 43 cm high×15 cm diameter stainless steel processing chamber is designed to allow for the deposition of thin films by sputtering and thermal evaporation, and for the cleaning of the surfaces by ion milling. The STM sample holder block is loaded into a Teflon sample basket which is fitted to a horizontal rotary/linear manipulator (see Fig. 1). This allows the sample holder block to be turned so that the sample faces any one of three ports on the top, front, or bottom of the chamber from which the different processing operations may be performed. The space in back of the sample basket is used for a viewport/door. The sample basket can also be used to manipulate tunneling tips. The chamber is equipped with a 240 l/s turbomolecular pump providing an ultrahigh vacuum environment ~10⁻⁹ Torr.

Since the processing chamber is directly coupled to the helium cryostat through a gate valve, contamination can be avoided by transferring the sample holder block and the tip without breaking vacuum. In order to transfer a tunneling tip onto the STM, the STM must be raised to the top of the cryostat (see below). The long horizontal manipulator can translate the basket to the cryostat top where the tip can be carefully pushed into a mating socket in the STM. In the case of transferring a newly prepared sample the maneuver is less...
A liquid-helium flow cryostat was chosen which enables the STM temperature to be varied either by helium gas flow (300–1.8 K) or by immersion in liquid helium (4.2–1.5 K). In order to reach the temperatures below 4.2 K, the sample tube of the cryostat is evacuated with a large 66 l/s mechanical pump (a somewhat smaller pump would still be adequate). Although the pump is only used when vapor is flowing or when the STM is immersed in liquid helium, a liquid nitrogen cold trap is installed in the pumping line to insure that no oil backstreams into the sample tube. The direct contact between the sample/tip and the helium gives a large cooling power which allows us to reach these low temperatures despite the multiple leads to the STM and the possible application of current through the sample.

At the heart of the system is a horizontally oriented, thermally compensated STM designed by Lyding et al. shown in Fig. 1(b). The design utilizes two concentric piezoelectric tubes, one for scanning and one for thermal compensation and inertial sample translation. Both the sample holder and the cut-away sample holder tube which support it are made of quartz to minimize thermal drift. The STM has a compact length of 3.8 cm easily fitting down the 5.7 cm diameter cryostat sample tube. The tube passes through the bore of an 8 T superconducting solenoid, the direction of the magnetic field is parallel to the scanning surface of the horizontally oriented STM. Our design would have to be modified for applications which require the field to be perpendicular to the surface. This could be accomplished by incorporating a vertically oriented STM or by using a horizontally oriented cryostat.

The compactness of the STM results in very high normal-mode vibration frequencies (greater than 20 kHz), which makes this design much less sensitive to room vibrations than larger STM’s. However, since our STM is directly coupled to the large cryostat, good vibration isolation is important to achieve a stable tunneling signal. We isolate the system from floor vibrations by resting it on four air springs inflated to 30 psi with 400 kg of lead bricks stacked onto the base. In order to eliminate vibrations from the mechanical pump (located 10 m away from the cryostat), the final vacuum connection is made to the system with a 1 m length of 2.5 cm inner-diameter rubber vacuum tubing. We have also found it helpful to reduce other acoustical noises in the lab by surrounding the system on three sides with acoustical damping panels.

The sample is brought into tunneling range by means of
inertial translation. This requires the sample holder block to slide along the groove in the sample holder tube with very little friction. For the purpose of electrical contact to the sample, the upper surface of the sample holder tube is metallized as are contact pads on the bottom of the sample holder. These metallized surfaces must be well polished and clean to insure that the inertial translation will work reliably during cryogenic operation. (Our cleaning routine includes an isopropyl alcohol wash and the application of a little MoS₂ powder).

The tunneling current is detected using a current-to-voltage preamplifier mounted to the top of the cryostat to minimize the effects of electromagnetic interference. Our custom-built electronics feature standard feedback and scan-control circuitry interfaced with a personal computer through a commercial data acquisition board. The output voltage to the STM's scanning piezoelectric tube (PZT-5H) has a maximum range of ±250 V yielding a scan range of 5 μm at 300 K and 0.6 μm at 1.5 K.

III. OPERATION

In a typical data run the cryostat sample tube is first purged with nitrogen gas and roughed out to a pressure of a few hundred milliTorr (with the gate valve shut). The sample holder is then loaded into the processing chamber and placed in the sample basket. By evacuating the processing chamber with the turbomolecular pump, the sample surface can be prepared in a UHV environment. Once the sample is ready, the gate valve is slowly opened so that the sample tube of the cryostat is evacuated. The sample holder is then transferred onto the STM and subsequently lowered with the STM down the sample tube to the cryostat bottom.

We have found that precooling the system while the sample tube is filled with an exchange gas causes unnecessary contamination of the sample surface. The problem is that contaminants drawn from the warmer stainless steel walls near the top of the cryostat are then cryopumped downward to the STM. The surface is kept much cleaner by containing the effects of electromagnetic interference. Our custom-built electronics feature standard feedback and scan-control circuitry interfaced with a personal computer through a commercial data acquisition board. The output voltage to the STM's scanning piezoelectric tube (PZT-5H) has a maximum range of ±250 V yielding a scan range of 5 μm at 300 K and 0.6 μm at 1.5 K.

FIG. 2. Demonstration of variable temperature operation on NbSe₂. (Left) Two 30 Å×30 Å topographic images showing atomic resolution at room temperature and additional CDW corrugations below the CDW transition temperature. (Right) Spectroscopic data showing the appearance of both a CDW gap (ΔCDW) and a superconducting gap (ΔSC) as the temperature is lowered.

Once the desired temperature has been reached, the STM can be used in three different ways to characterize the surface. The topography is obtained by monitoring the feedback signal necessary to keep a constant tunnel current (hence tip–sample separation) while rastering the tip location over a rectangular area. In addition to identifying prepared structures on the surface, the topography can also reveal the atomic lattice, which provides a good test of the tip quality and surface cleanliness. Second, the STM can be used in a scanning spectroscopy mode which produces an image of the surface representing the differential conductance dI/dV at a particular voltage. (The differential conductance is a significant quantity since it is directly related to the local electronic density of states.) This is accomplished by superimposing a small sinusoidal voltage with the sample bias voltage, resulting in an ac tunneling current proportional to dI/dV which is then fed into a lock-in amplifier. The lock-in output is monitored while the tip is rastering to produce the spectroscopic image. Last, the STM can produce dI/dV(V) by interrupting the scan and holding the tunneling tip fixed (usually for about 30 s) while ramping the bias voltage (sinusoidal voltage still superimposed). Although this procedure yields the most detailed electronic information, it also demands the most stability as a constant tip–sample separation must be maintained to a precision better than 0.1 Å without the use of feedback. By repeating the procedure at different locations, dI/dV(V,r) can be completely mapped out on the surface.

We have found our system to be able to obtain high-quality topographical and spectroscopic data at room temperature and cryogenic temperatures. Variable temperature operation is demonstrated in Fig. 2 on a NbSe₂ sample. NbSe₂ undergoes two phase transitions: a charge density wave (CDW) transition at 33 K, and a superconducting tran-
FIG. 3. (a) $dI/dV(V,r)$ on the surface of NbSe$_2$ at 1.6 K showing the expected homogeneous behavior. (b) A representative curve normalized to the value at high voltage. The superconducting energy gap in the density of states is clearly seen as the pronounced dip around zero voltage. The solid curve is a BCS fit with the parameters as indicated.

The remaining figures show data from a sample prepared in situ which consisted of a large NbSe$_2$ crystal onto which 7 Å of Au were deposited. Figure 4 shows a topographical image of the sample surface obtained at 4.2 K. The periodic structure is the exposed layer of Se atoms while the deposited Au gives rise to the irregular structure on the bottom left. In Fig. 5 a $dI/dV(V)$ taken with the tip positioned on one of these Au islands is compared to that of bare NbSe$_2$. The structure observed inside the energy gap arises from discrete quasiparticle bound states in the Au (a normal metal). This experiment represents the first local observations of discrete bound states in an artificially prepared structure.

In summary, we have constructed and tested a variable-temperature STM system featuring in situ sample/tip processing. The processing takes place in a UHV chamber vacuum coupled to liquid helium flow cryostat. High-quality topographical and spectroscopic data are obtained using a Lyding-design variable-temperature STM mounted to a motor-driven threaded rod. When lowered to the bottom of the cryostat, the sample temperature can be varied from room temperature to 1.5 K by helium gas flow or immersion in liquid helium.

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3In addition to our own experience this phenomenon has been reported by M. A. Dubson and J. Hwang, Rev. Sci. Instrum. 63, 3643 (1992).
4For an introduction to UHV sample preparation techniques and a brief overview of UHV STM see H. Lüth, Surfaces and Interfaces of Solids (Springer, Berlin, 1993).
5Purchased from Janis Research Co., 2 Jewell Drive, Wilmington, Mass. 01887, (508) 657-8750.
8 A vertical piezoelectric inertial walker such as the one developed by Ch. Renner, Ph. Niedermann, A. D. Kent, and Ø. Fischer, Rev. Sci. Instrum. 61, 965 (1990), could be modified to allow for a removable sample holder.
10 Ithaco model #564 purchased from Ithaco Inc., 735 W. Clinton St., Box 6437, Ithaca, NY 14851, (607) 272-7640.