Integrated cryogenic scanning tunneling microscopy and sample preparation system

S. H. Tessmer and D. J. Van Harlingen
Department of Physics and Materials Research Laboratory, University of Illinois at Urbana–Champaign, Urbana, Illinois 61801

J. W. Lyding
Department of Electrical and Computer Engineering, University of Illinois at Urbana–Champaign, Urbana, Illinois 61801

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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[1] (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.[2] 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.[3] 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 $\sim 100\ \mu$V, 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$\times$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.[4] 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 $\sim 10^{-9}$ 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

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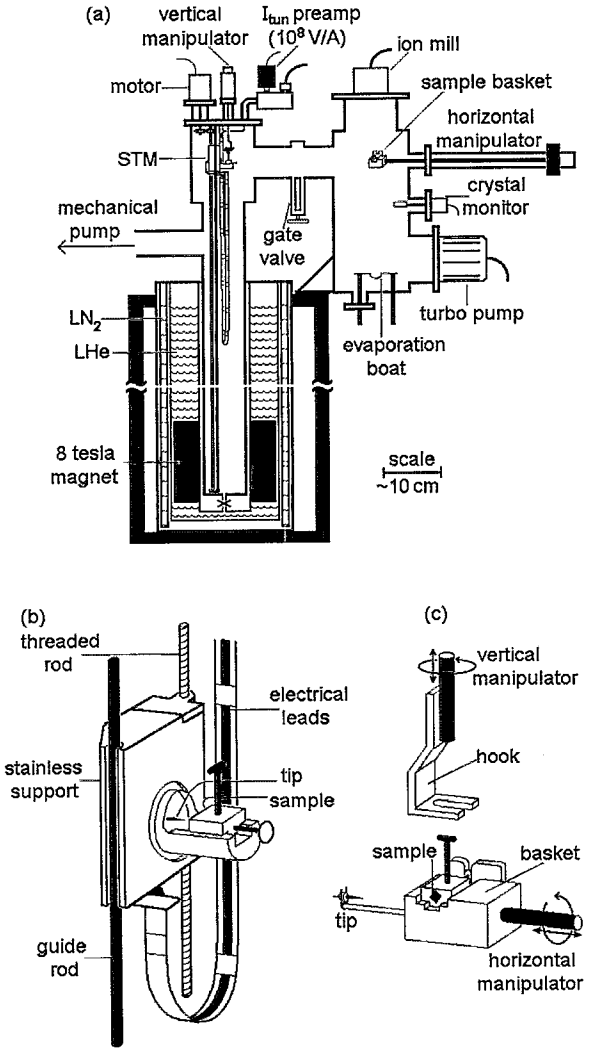

FIG. 1. (a) Sketch of our cryogenic STM system drawn roughly to scale. The system features a liquid-helium flow cryostat with an adjacent processing chamber allowing for *in situ* modification and cleaning of the sample and tip. (b) An enlargement of the STM and the vertical translation assembly. (c) An enlargement of the fittings used for sample manipulation.

delicate, although an extra step is involved due to the orientation of the sample holder. The sample basket is first positioned directly beneath a second rotary/linear manipulator at the top of the cryostat. This manipulator is equipped with a hook that can latch on to the sample holder so that it can be lifted off the basket, rotated 90° (the sample now facing the tunneling tip), and lowered onto the STM.

The STM is mounted on a captive acme threaded rod which is coupled to a DC gear motor via a direct-drive feedthrough (see Fig. 1) so that it can be moved vertically throughout the sample tube of the cryostat. The electrical leads (12 low-heat-leak stainless-steel coaxial cables) are tethered to a phosphor-bronze support strip which constrains them to follow the STM's motion in a controlled way. Introducing a curvature across the width, like in a tape measure, to a 1.3 cm×0.16 mm×150 cm strip of this nonmagnetic, low thermal-conductance alloy gives it a tendency to stay straight along its length. Such a strip is attached to the STM support block forming a loop that drapes beneath the STM, with straight sides and a well defined curve at the bottom, as shown in Fig. 1(b). We attach the electrical leads to this strip by a wrapping of Teflon tape every few cm so that it easily slides along the inside bore of the sample tube. As the STM is moved downward from the top position, extra length of the leads is made available from the loop of decreasing size underneath the STM. In this way, the STM with newly prepared sample and tip can be lowered to the bottom of the cryostat. Once the system has been cooled, however, we find that the STM must remain stationary because the insulation in the low-temperature leads becomes too stiff to bend around the loop without cracking.

A liquid-helium flow cryostat[5] 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.*[6] 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;[7] 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[8] 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.[9]

The sample is brought into tunneling range by means of

inertial translation.[6] 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 MoS_2 powder).

The tunneling current is detected using a current-to-voltage preamplifier[10] 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[11] 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.

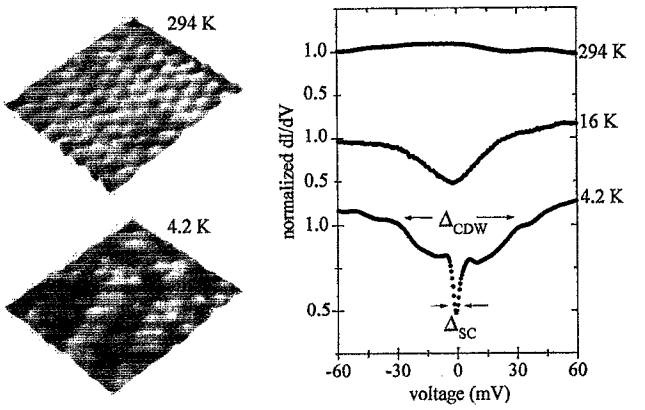

FIG. 2. Demonstration of variable temperature operation on $NbSe_2$. (Left) Two 30 Å×30 Å topographic images showing atomic resolution at room temperature and additional CDW corrugations below the CDW transition temperature. (Right) Spectroscopic data showing the appearance of both a CDW gap (Δ_{CDW}) and a superconducting gap (Δ_{SC}) as the temperature is lowered.

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 continuing to evacuate the sample tube with the turbo pump as liquid nitrogen is introduced into the cryostat reservoirs (with the needle valve connecting the sample tube to the helium reservoir shut). This allows the STM to precool by radiation in a high-vacuum environment $\sim 10^{-8}$ Torr. The resulting low temperature gradient (0.2 °/min) minimizes thermal-contraction stresses on the STM which can significantly shorten its operating lifetime.

When the STM reaches around 150 K, liquid helium is introduced into the cryostat. The gate valve is then shut and the turbo pump switched off as further cooling is achieved by helium vapor rising through the needle valve at the bottom of the sample tube. Again to avoid high thermal stress on the STM, the vapor flow rate is adjusted so that cooling proceeds at ~0.7 °/min. Therefore, we take about 16 h in total to cool a newly prepared sample to liquid helium temperatures. For temperatures at or below 4.2 K, we have achieved the best results operating the STM immersed in liquid helium. This environment provides excellent temperature stability while freezing out any residual contaminants. In practice, tunneling through liquid helium (an insulator) is much like vacuum tunneling. Due to the long cooling time, the system is not suitable for atomic-resolution applications involving reactive surfaces. However, by the use of samples which are chemically inert such as graphite, $NbSe_2$, or noble metals, the cleanliness of the surface can be maintained allowing atomic resolution (see for example Figs. 2 and 4).

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 scanning 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, $dI/dV(V,\mathbf{r})$ 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 $NbSe_2$ sample. $NbSe_2$ undergoes two phase transitions: a charge density wave (CDW) transition at 33 K, and a superconducting tran-

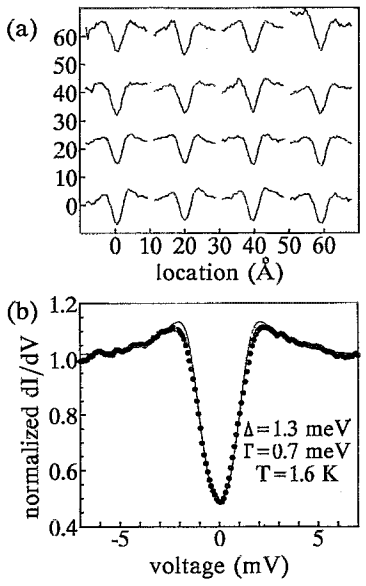

FIG. 3. (a) $dI/dV(V,r)$ on the surface of $NbSe_2$ at 1.6 K showing the expected homogeneous behavior. (b) A representative curve normalized to the value at high voltage. The superconducting energy gap in the density of states is clearly seen as the pronounced dip around zero voltage. The solid curve is a BCS fit with the parameters as indicated.

sition at 7 K. The charge density wave with a period of approximately three atomic spacings is apparent at 4.2 K in Fig. 2 (left). The spectroscopic curves shown in Fig. 2 (right) display features of both the CDW gap and the superconducting gap. Figure 3(a) shows $dI/dV(V,\mathbf{r})$ for several locations on the surface of superconductor $NbSe_2$. As expected, the superconducting energy gap is observed everywhere on this homogeneous sample. This demonstrates the stability of our system at cryogenic temperatures and the reproducibility of the spectroscopic measurements. Figure 3(b) shows a representative curve with a BCS fit. A nonzero depairing parameter Γ results from the high current density inherent to the STM.[12]

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.[13]

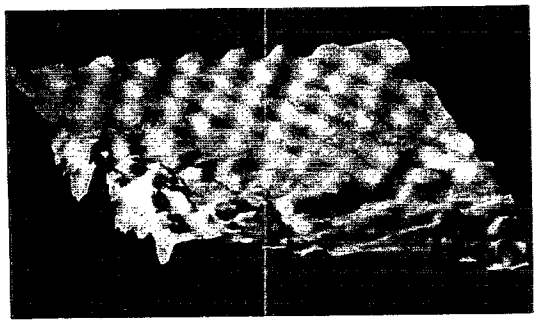

FIG. 4. A 30 Å×30 Å topographic image obtained at 4.2 K of a $NbSe_2$–Au sample prepared in our system. Both the atomic structure of $NbSe_2$ (top) and irregular topography indicative of Au (bottom) are present.

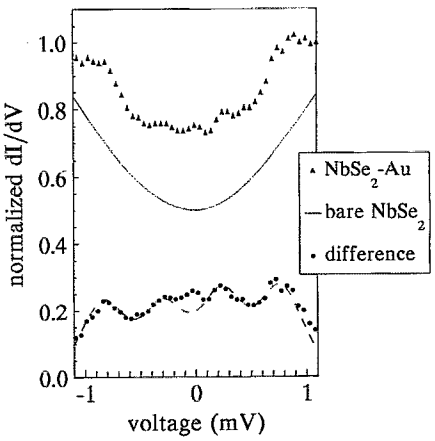

FIG. 5. A comparison between a dI/dV obtained on a 6 Å tall Au island of the $NbSe_2$–Au sample and a representative dI/dV of bare $NbSe_2$. The structure found in the spectra of the $NbSe_2$–Au sample arises from quasiparticle bound states localized in the Au. The dashed curve is a theoretical fit based on a simple bound state model.

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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