Chapter 3

Experimental setup of the low temperature STM

This chapter describes the ultra-high-vacuum (UHV) LT-STM developed during this thesis and used for the measurements presented here. The LT-STM, built with the help of Christian Roth, is based the design of Gerhard Meyer [67] and was further optimized by the members of our research group [68, 69].

Because of the growing research interest in scanning tunneling microscopy as a powerful tool for studying single atoms and molecules on surfaces, much effort has been put in the past decades to design STMs that are able to reach atomic resolution and to manipulate atoms and molecules, as well as to perform spectroscopy measurements [70–73]. In order to fulfill these tasks, the lateral resolution of the STM must be less than the interatomic spacing of a crystal surface, typically around 2 to 3 Å, while the vertical resolution must be around 0.01 Å. In fact, due to the exponential I(z) relationship in a STM junction (see Equation 2.20), a change in the tip-sample separation of one Angstrom results typically in about an order of magnitude variation in the tunneling current. Furthermore, mechanical vibrations of the system and the thermal drift must be extreme small to allow the tip to stay stable above a single atom or molecule for hours.

The system described here is characterized by an inherent mechanical stability and good drift compensation. It operates at variable temperatures between 5 and 300 K.

3.1 Vacuum system

The ultra-high-vacuum chamber is divided into two parts. One is used for the preparation of the sample (see left side of Fig. 3.1), while the other contains the cryostat with the STM. The two chambers are separated by a 150 mm UHV-gate-valve and can be separately pumped. The pumping system consists of a turbo molecular pump (260 l/s) connected to

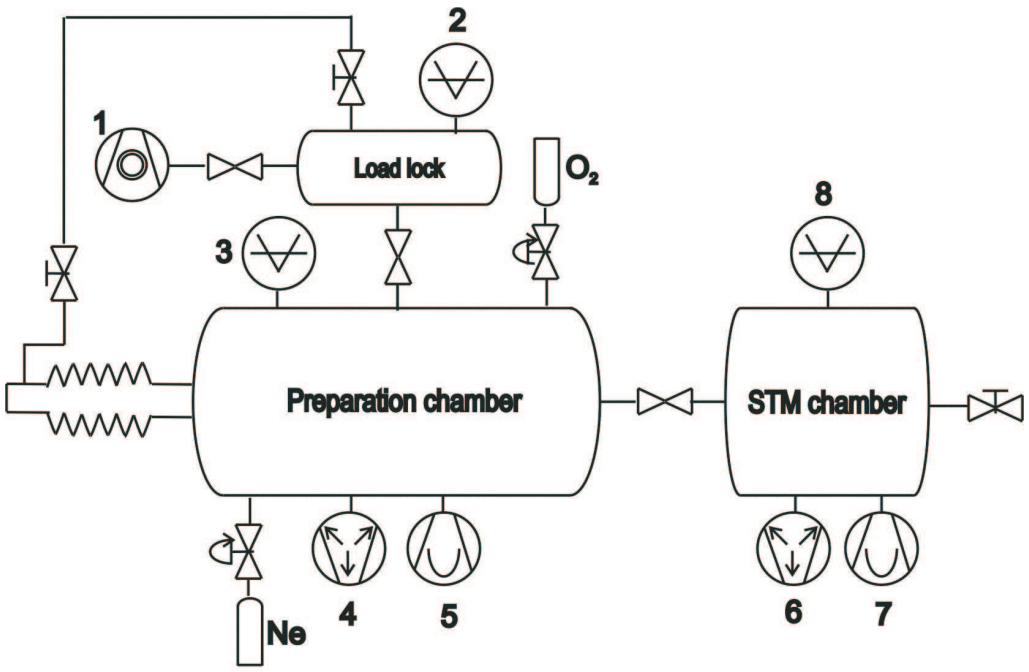

Figure 3.1: Pumpimg system: (1) Turbo molecular pump (260 l/s). (2) Pressure gauge (full range). (3) Ionization gauge. (4) Ion getter pump (50 l/s). (5) Titanium sublimation pump. (6) Ion getter pump (400 l/s). (7) Titanium sublimation pump. (8) Ionization gauge.

the preparation chamber through a load lock chamber; two titanium N_2 cooled sublimation pumps (one per chamber), and two ion getter pumps (50 l/s and 400 l/s, again one per chamber). The base pressure in the preparation chamber is in the upper 10^{-10} mbar range. The cryostat itself operates as a cryo-pump, allowing pressures lower than 10^{-12} mbar. Due to this extremely low pressure, the sample surface remains clean for several weeks. Ionization gauges (Varian) measure the pressure in the STM and preparation chambers, while a full range gauge (Pfeiffer) is used in the load lock chamber.

3.2 The preparation chamber

The preparation chamber (see Fig. 3.2) is equipped with a mass spectrometer (Pfeiffer), an ion sputter gun (Varian), gas inlets for neon gas (used for sputtering) and oxygen gas for the preparation of oxide films. An evaporator (Omicron) is used for the deposition of metals (vanadium in this work) and a home-built evaporator for depositing molecules. A manipulator (VAb) is utilized to transfer the sample to the STM chamber. A load lock chamber with a magnetic transfer bar facilitates the exchange of samples in and out of the

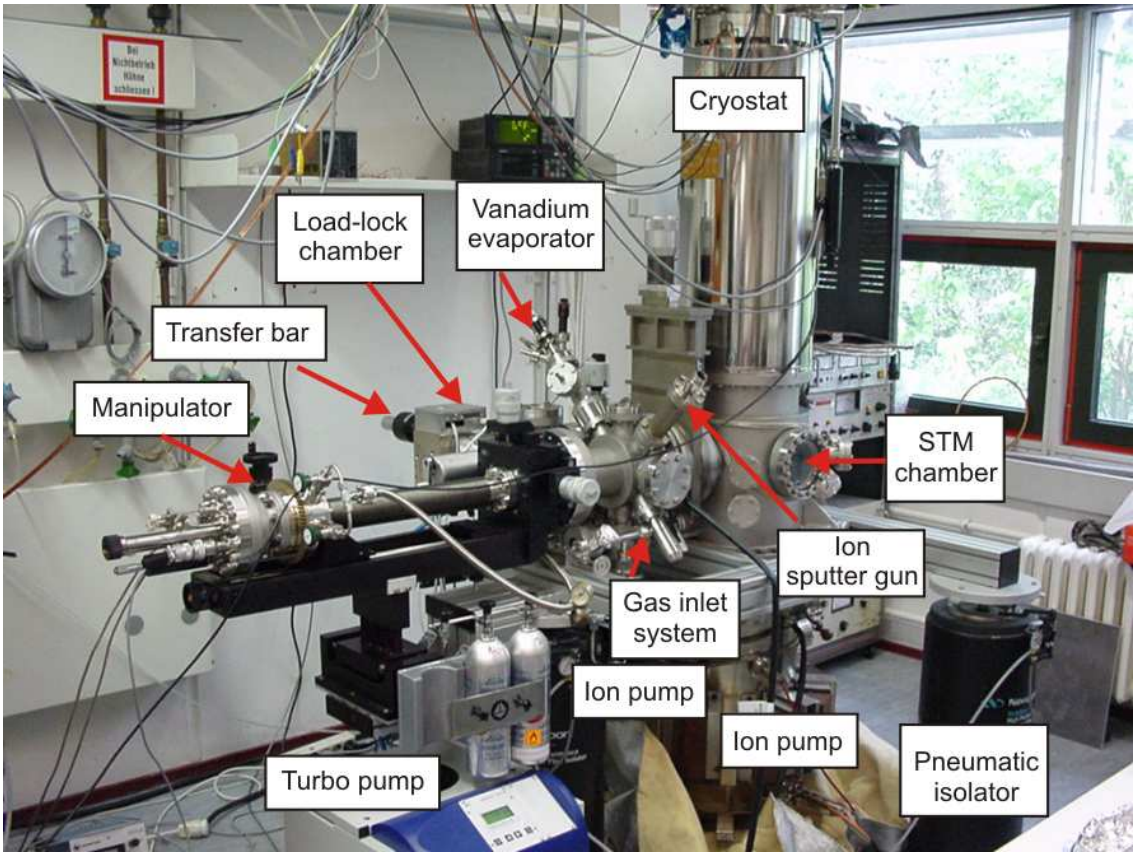

Figure 3.2: Photograph of the UHV chamber constructed.

UHV environment without venting the entire preparation system. The main parts of the preparation chamber and sample holder will be presented in the following:

- The manipulator is used to move the sample during the preparation steps and for transferring it into the STM. It can be moved in all three spatial directions and can be rotated around its axis. A T-bar mounted on the end of the manipulator (see Fig. 3.3(a),(b)), is utilized to fix the sample holder. On the manipulator back part (see Fig. 3.3(c)), contacts pins are mounted for electrical contact, providing for heating the sample and measuring its temperature. The top end of the manipulator can be cooled with liquid nitrogen or liquid helium to reduce the temperature of the sample before it is transferred in the STM. If liquid helium is used for cooling, about 30 min are needed to cool down the sample from room temperature to around 40 K. The temperature of the manipulator end is measured by a type K thermoelement.

- In the UHV evaporator, vanadium is evaporated from a rod heated by electron bombardment. The electrons are emitted from a heated filament and accelerated towards

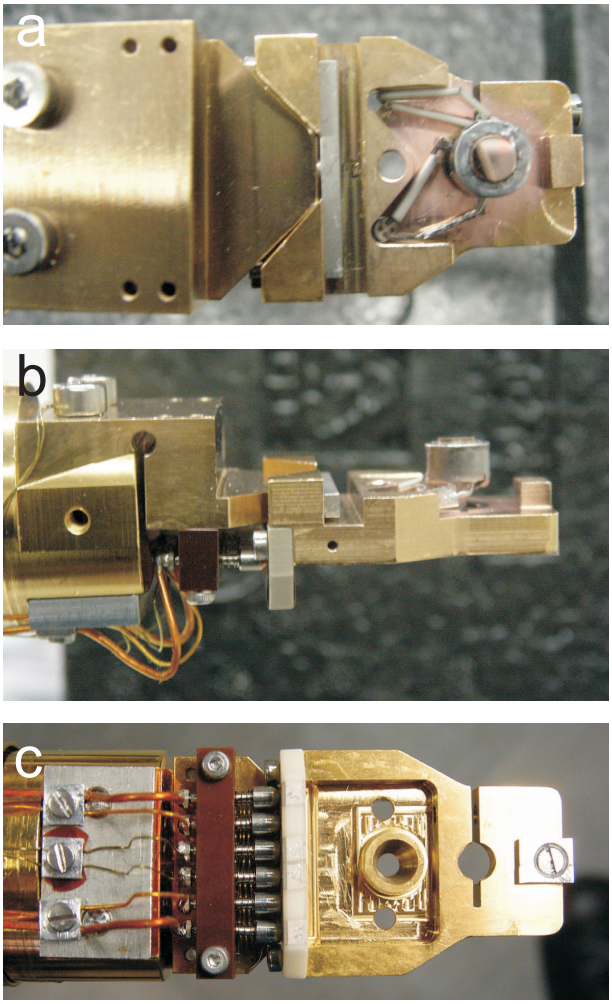

Figure 3.3: (a) Top view of the manipulator end with the T-bar grabbing the sample holder. (b) Side view of the manipulator like in (a). (c) Back part of the manipulator. It is possible to see the wires used for sample heating (external cables) and the thermoelement cables (at the center) for measuring the sample temperature. The contacts between manipulator and sample holder are stabilized by home made elastic contact pins.

the vanadium rod by a high voltage. The evaporator is contained in a water-cooled cell which guarantees a localized heating at the apex of the rod only. Furthermore, it ensures a low basis pressure. An ion collector serves as a flux monitor at the beam exit. At a different electron emission current and e-beam voltage, the flux of evaporated atoms is directly proportional to the ion flux. Nevertheless, to exactly calibrate the amount of vanadium evaporated onto the sample, a quartz crystal micro balance is installed directly under the sample.

- The evaporator used for depositing molecules is home built. Its design (shown in Fig. 3.4) is very simple, and its geometry allows it to be mounted in the load lock chamber. It consists of a resistively heated crucible with a small hole at the top. The crucible temperature is measured with a type K thermocouple. A quartz microbalance, attached at a distance r = 43 mm from the source, is used to measure the molecular flux. Its plane is tilted 45° with respect to the connecting crucible and

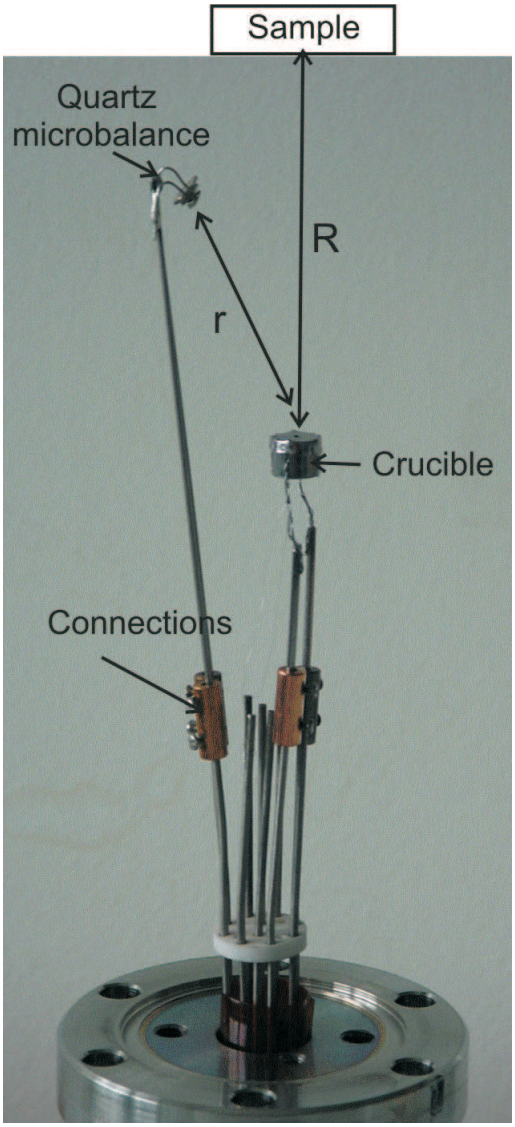

Figure 3.4: Photograph of the molecular evaporator. Its components are indicated in the photo. The distance between source and quartz micro balance is r $\approx$ 43 mm, while the distance R between source and sample is 280 mm.

sample. Under such a geometry, the quartz microbalance is working in an excellent way, without showing temperature effects during the evaporation of the molecules.

- The sample is mounted on a copper (gold plated) sample holder, shown in Fig. 3.3(a). It can be grabbed by the manipulator to transfer it from the preparation chamber into the STM. In its back part, it has a ceramic plate covered by conducting pads. These pads form electrical contacts to the manipulator for heating the sample surface and for measuring its temperature. Besides, when the sample is inside the STM, they allow to apply the bias voltage. The sample is mounted on an button heater in order to be able to heat the surface during its preparation. The temperature of the sample is measured with a type K thermocouple.

3.3 Cryostat

The constituents of the low temperature system are schematically indicated in Fig. 3.5. The STM is cooled to about 5 K by a He bath cryostat system (Cryovac). It consists of two tanks: the inner one (with a volume of 8.5 liters) is filled with liquid helium, while the outer (with a volume of 18 liters) is filled with liquid nitrogen. The nitrogen tank serves as a radiation shield and decreases the He consumption. To improve the thermal isolation of the system, an additional radiation shield cooled by cold exhaust helium gas, is mounted in between the tanks. The temperature of the cryostat is measured with a Si-diode (Lake Shore).

The microscope is surrounded by two thermal radiation shields of pure aluminium, one contacted to the helium tank and the other one to the nitrogen tank. These radiation shields are used to thermalize the scanner at liquid helium temperature and act as cryo-pumps in the STM chamber. Aluminium is chosen because of its good thermal conductivity and low emissivity. Shutters in the shields allow the sample transfer in and out of the STM. They can be opened and closed by means of a rotary feed-through, shown in Fig. 3.5. To reduce helium consumption, the shutters are cooled by copper braided wires that link the shutters to the radiation shields. A linear feed-through mechanism (visible in Fig. 3.5) is used to pull the scanner in contact with the cooling plate at the bottom part of the He shield, allowing the cooling of the scanner to helium temperature in about 3 hours.

To permit optical access through the shields, two windows (DURAN) have been installed. With the help of an optical microscope one can magnify the tip sample junction, thus a well-controlled tip approach can be achieved. Furthermore, one can observe and control the sample transfer in the STM.

With the described cryogenic system, the temperature of the STM remains stable at 5 K with a consumption rate of less than 0.07 l He/h after the cool-down process. Once the STM is at 5 K, helium has to be refilled every 5 days. However, since the nitrogen tank is empty after 44 hours, continuous measurements can only be performed for 44 hours.

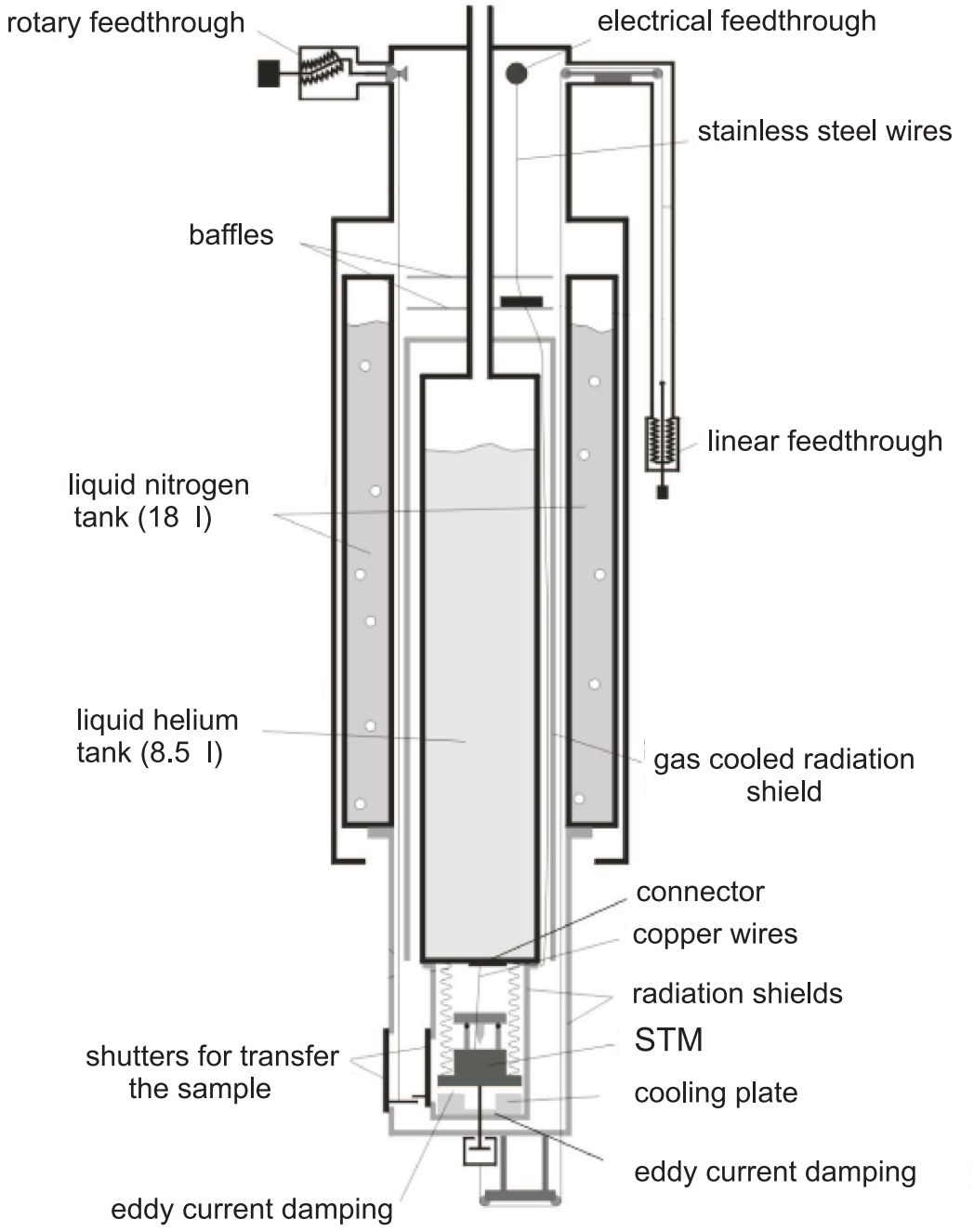

Figure 3.5: Schematic of the bath cryostat system from reference number [68]. Its components are indicated in the image.

3.4 Scanner

The scanner in Fig. 3.6, is of a Besocke beetle type [74, 75]. It was designed by Gerhard Meyer. Its components are all made of the same material (copper), thus avoiding different temperature expansion coefficients. In addition, they are all gold plated to reflect light and reduce the emissivity. The scanner is compact, to enable good thermal contacts between components and for being less sensible to vibrational noise, stiff and small, so that its eigenfrequency is high and does not couple to external vibrations. Moreover, Besocke scanners allow to *scan* surfaces as well as to *approach* the tip in a very controlled manner towards the sample from macroscopic distances to tunneling conditions and to *position* the tip macroscopically over the sample.

As shown in Fig. 3.6, the scanner is composed of three lateral piezoelectric tube elements (Staveley Sensors, material EBL No. 2). Those hold, through three sapphire balls, a base copper plate called *approach ring*. Another piezoelectric tube supporting the tip is mounted in the center of the approach ring.

To perform a *scan* parallel to the surface (x- and y-directions), one uses the lateral piezoelectric tubes, while for the regulation of the tip-surface distance (z-direction) the central piezoelectric tube is used.

Actually, one could also use the central-piezo for scanning in all x-,y-,z- directions. However, the scanning distance is much smaller in this case. For x-,y- motion, one applies a potential ramp separately to each lateral piezoelectric tube $\pm$ x and $\pm$ y, where voltages of opposite polarity are applied to the opposite ($\pm$) piezo-elements. In this way, the three piezos will move synchronously in the same direction and the tip, together with the approach ring, will follow this movement.

The *coarse approach* is necessary, since the tip is usually positioned far away from the sample during the idle time of the microscope. Thus, one avoids destruction of the sample by a tip crash induced by accidental movements. The coarse approach is done using an inertial sliding technique. By applying voltages having a sawtooth form to the outer tubes, small cycles of stick and slip movements of the ring relative to the sapphire balls are performed. If all three tubes are driven to move tangentially as shown in Fig. 3.7, the approach ring will perform a macroscopic rotation by summing up the tiny stick and slip movements.

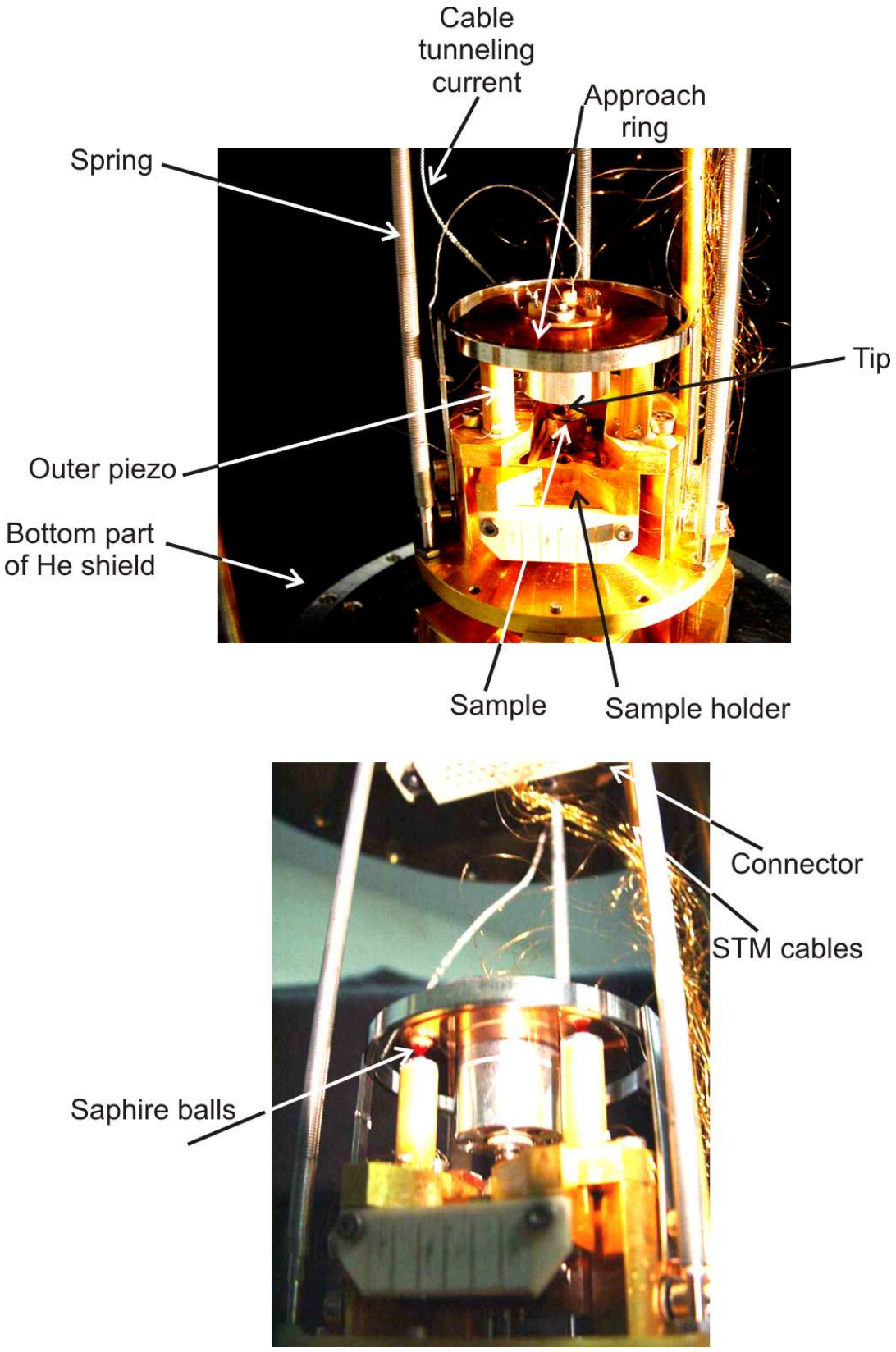

Figure 3.6: Photographs of the microscope. The components of the scanner are indicated in the images.

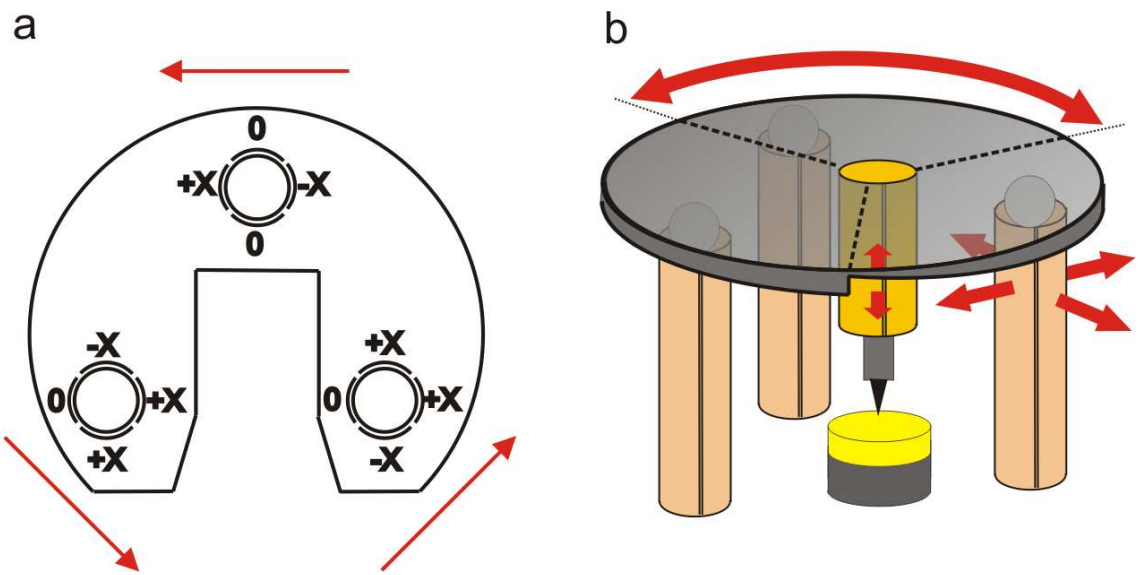

Figure 3.7: Schematic of the coarse approach: when the outer piezos are tangentially moved by applying voltages with a sawtooth form (indicated by $\pm$ X and where the maximal applied voltage is $\pm$150 V) as shown in (a), the approach ring rotates. Since the approach ring is not flat this results in vertical movement of the tip (b).

Since the approach ring is not flat, but has three ramps (every 120° with 2° inclination), this rotation is accompanied by a perpendicular motion of the tip. With our ramp, the maximal vertical approach distance of the tip is 0.5 mm. It is important to note that the *approach ring* has to be carefully polished to avoid uncontrolled slidings.

By applying a sawtooth signal to the outer tubes, one can also move the outer piezos along a common direction. In this way the lateral *positioning* of the tip on the sample is performed.

During scanning, coarse approach, and positioning the piezo segments have to be addressed with the different voltage signals. An external switch box facilitates the switching between rotation and lateral motion of the STM tip under computer control. The form and the frequency of the voltage pulses applied to the *approach ring* is optimized for each system in order to obtain a well controlled movement of the STM tip. At 5 K, the lateral voltage dependent elongation of our piezoelectric tube is about 50 Å/V, while at room temperature it is about 125 Å/V. The high voltage applied to the piezos is $\pm$150 V. This corresponds to a scan range of about 1.5 μm at low temperature and about 4 μm at room temperature. The lateral resolution of our microscope is in the order of 1 Å, while the vertical resolution 1 pm.

In the STM described here, the bias voltage is applied to the sample while the tunneling current is measured on the tip. The contacts for the bias voltage to the sample are of the same kind as the pins of the manipulator, visible in Fig. 3.3. A Si-Diode (LakeShore) is mounted on the scanner for measuring its temperature. Furthermore, a Zener diode (type BZY93C75), attached to the microscope, can be used for heating the scanner.

3.5 STM-Tip

Initial test measurements with a highly ordered pyrolytic graphite (HOPG) sample and PtIr tips have been performed in air. This material is often used for STM tips operating in air because of the inert character of Pt and the hardness of Ir. PtIr-tips have been obtained by manually cutting a PT-Ir wire (0.25 mm diameter) without further processing.

However, in all the measurements presented in this thesis, a tungsten tip has been used. It was prepared by electrochemically etching a tungsten wire (0.25 mm diameter) in a NaOH solution. Tungsten has been chosen because of its hardness, since contact between tip and sample can accidentally occur during the scanning.

After the preparation, the tip has been mounted on a magnetic (CoSm) holder at the end of the central piezo. This system allows to exchange the tip in the vacuum chamber. However, during this work a tip exchange has never been performed. In fact, by working with metal surfaces, there are several methods to increase the tip resolution changing the microscopic shape of the tip. Notice that for obtaining atomic resolution on relatively flat surfaces, the tip should terminate with a single atom but its macroscopic shape is of little importance.

One of those "tip-sharpening" methods "cleans" the tip by gently crashing it into the metal surface. After that, the tip is expected to be covered by metal surface atoms. Another technique to reform the tip-apex uses field emission of electrons under voltage pulses of ± 9 V. Finally one can apply high dc voltages (± 100 V) to the sample while bringing the tip in contact to the sample. This treatment is performed in a sample region far away from the measurement zone, because it damages the surface region close to it.

3.6 Wiring system

The wiring system is divided into three parts: wires in the microscope, in the cryostat and outside the chamber.

- In order to enhance the thermal conductivity between STM and cryostat, Kapton coated copper wires have been utilized for connections in the STM. Since wires connected directly to the STM should not transmit vibrations, thin flexible wires (ø0.8 μm) have been used. In addition, they have been anchored at various stages in the STM. The STM wires end on a connector (Hostrad) positioned at the bottom of the cryostat, allowing for dismounting the STM out of the cryostat without disruption of any cables. The wire for the tunnelling current is a shielded stainless steel cable.

- The wires from the connector to the end of the cryostat are made of shielded stainless steel wires. This material is chosen to reduce the thermal conductivity and minimize the heat transfer into the microscope by the cables. The cables pass in feed-through tubes through the cryostat. The tunnelling current amplifier is connected directly to the exit of the vacuum chamber.

- Outside the vacuum chamber, coaxial BNC cables have been used. The isolations and the grounding of the cables is very important for electrical noise isolation, as it will be explained in the Sec. 3.8.

3.7 Electronics

The STM electronics has been designed by Gerhard Meyer and Sven Zöphel [68]. It is as crucial as the mechanical parts of a STM. The system allows a complete digital control of the imaging process. A DSP (digital-signal-processor) is connected with the analog electronic by four D/A and four A/D (digital to analog and analog to digital) converters. One D/A supplies the bias voltage (between +10 V to -10 V with 1 mV resolution). The other three D/A converters use high-voltage amplifiers to provide the voltage to the piezoelectric tubes. An A/D converter reads the amplified tunneling current into the DSP. The amplifier (Femto) can operate with a gain between 10^3 and 10^{11} V/A at a typical bandwidth of 7 KHz in normal STM operation.

The measured and amplified tunneling current is compared to the set point current (typically in the range of nA) and the resultant error signal is fed into a feedback loop, which consists of a proportional (P) amplifier and a digital integrator (I). The feedback signal is finally fed into a high-voltage amplifier which generates the amplified voltage signal applied to the z- piezoelectric drive. The DSP-programm transmits the measured data to another program in a PC.

The STM can be operated either in the *constant current* mode or the *constant height* mode. In the constant current mode, the feedback loop adjusts the height of the tip during scanning so that the tunneling current between the tip and the sample is kept constant. The height adjustment is performed by applying an appropriate voltage V_z to the z piezoelectric drive while the lateral tip position (x,y) is determined by the corresponding voltages V_x and V_y applied to the x and y piezoelectric drives. Alternatively, in the constant-height mode, the feedback loop is switched off and the modulation of the tunneling current is measured while the tip scans at a constant height over the surface.

3.8 Isolation

Noise isolation is one of the main challenges in constructing a LT-STM in order to guarantee a high stability and good resolution during measurements. The noise can be mechanical or electrical. In the following, I will discuss the solutions used to minimize these external noise sources.

3.8.1 Mechanical noise

Mechanical noise originates from external building vibrations as well as from acoustic waves. External mechanical oscillations are typically in the range of 1 to 100 Hz. To insulate the system from such mechanical disturbances, pneumatic suspension legs are used. In particular, three pneumatic isolators (Newport) with resonance frequencies below 2 Hz support the whole system. Another isolation step is obtained by hanging the STM-scanner on spiral springs connected to the lower part of the liquid-helium bath cryostat. These springs have an eigenfrequency of about 2 Hz. To reduce oscillations of the springs, magnets are mounted at the bottom of the microscope, which operate as eddy current damping devices.

The helium tank itself is suspended on a small tube which serves for the helium refill.

Such a system can oscillate like a pendulum. It is possible to control the alignment of helium tank by means of a screw system at the top of the cryostat. The oscillations of the pendular system are damped by means of magnets mounted at the bottom of the helium tank shield.

To prevent further vibrational coupling, the first resonance frequency of the STM-scanner unit should be as high as possible, and the number of vibrational modes should be small, which means that the construction of the STM unit has to be compact and rigid. Vibrational noise can also arise from the motion of the wire carrying the tip current. This issue has been addressed by carefully clamping all of the STM wires in place.

Finally, to achieve the maximal possible stability, the mechanical pumps are switched off during measurements, and all connectors that are not used for the STM measurements are removed.

3.8.2 Electronic noise

The tunneling current is very small on the order of nA to pA. Therefore, it is highly vulnerable to electronic noise from induction of electromagnetic waves in the laboratory. Thus, the cable carrying the tunneling current is carefully shielded. Furthermore, the weak tunneling current signal is amplified and filtered immediately at the exit of the vacuum chamber. A more delicate electromagnetic isolation is achieved by accurately grounding the STM. Many BNC cables and different electronic instruments are connected to the STM, each of them has its own ground. Such a situation is very inconvenient for low noise measurements. To overcome this problem, the shielding of all the cables is designed so that there is no overlapping ground. The cable of the ion getter pump is used as the STM ground. In addition, the STM chamber is connected with the electronic components by a thick copper wire. To further decrease the sources of electrical noise, nonessential electrical devices are switched off during measurements, and all the connections that are not used for the STM measurements are removed.

3.9 Performance of the LT-STM

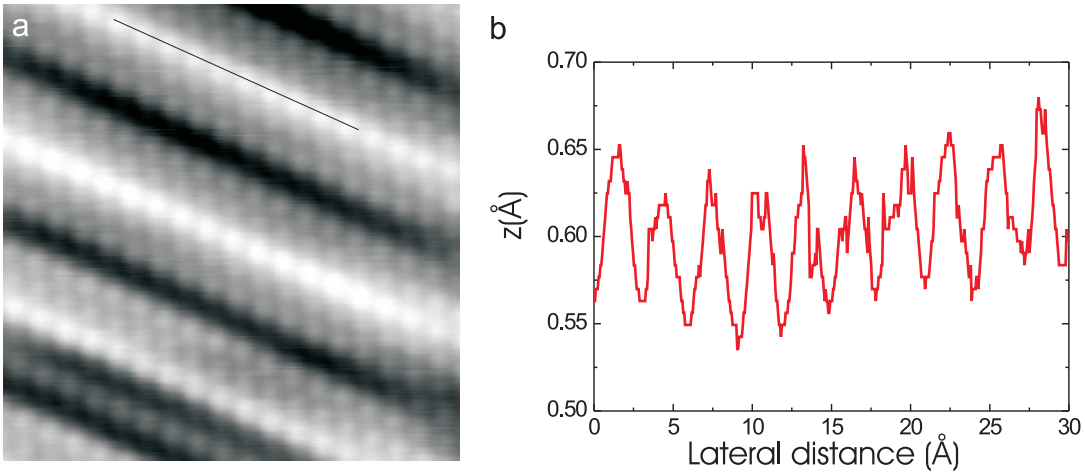

Figure 3.8: (a) Atomically resolved STM image of the Au(100) (5×2) surface reconstruction. STM parameters: U = 0.09 V, I = 40 nA, T = 5 K. Image dimension = (50×50) Å^2. (b) Line scan along the line indicated in (a), showing the atomic corrugation.

The high spatial resolution of the LT-STM built during this thesis work is documented in many atomic resolution images. An example is shown in Fig. 3.8(a), where an Au(100) surface is scanned with atomic resolution. Notice that the strong corrugation in the image is due to a (5×2) surface reconstruction [76]. Atomically resolved images are used to calibrate the x-,y-piezo-constants. Furthermore they exhibit the crystal orientation of the surface. The z-piezo constant is calibrated using surface steps. Since piezo constants are temperature dependent, a calibration is valid only at the temperature at which the atomically resolved image is taken. A line scan performed along the [110] direction is shown in Fig. 3.8(b). The atomic corrugation parallel to the surface row amounts to about 0.09 Å, while the noise level is about 0.03 Å.

The stability of LT-STM is proven by scanning spectroscopy measurements. An example of a dI/dV spectrum recorded on a Au(111) surface is shown in Fig. 3.9.

It is known, that standing wave patterns are formed in the LDOS of metals exhibiting Shockley-type surface states, when surface state electrons are scattered by surface defects on metal surfaces [77–79]. These standing waves can be observed with LT-STM by scanning at low voltages [80]. Moreover, surface states on noble metals surfaces can be detected by scanning tunneling spectroscopy measurements, because at the energy of

the surface state band gap, electrons can tunnel into the surface state and the tunneling conductance (dI/dV signal) exhibits a strong enhancement [81]. In the spectra presented in Fig. 3.9 a step like peak is observed in the conductance at U = -510 mV below the Fermi energy, which corresponds to the surface state of Au(111).

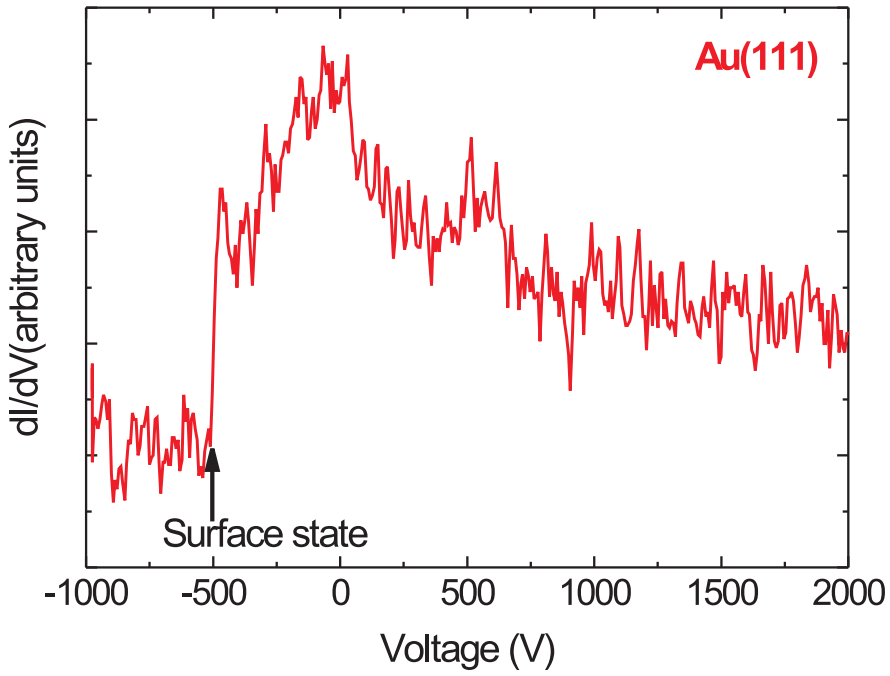

Figure 3.9: STS spectra on Au(111) where the surface state signature is visible as a step at 510 mV below the Fermi energy.