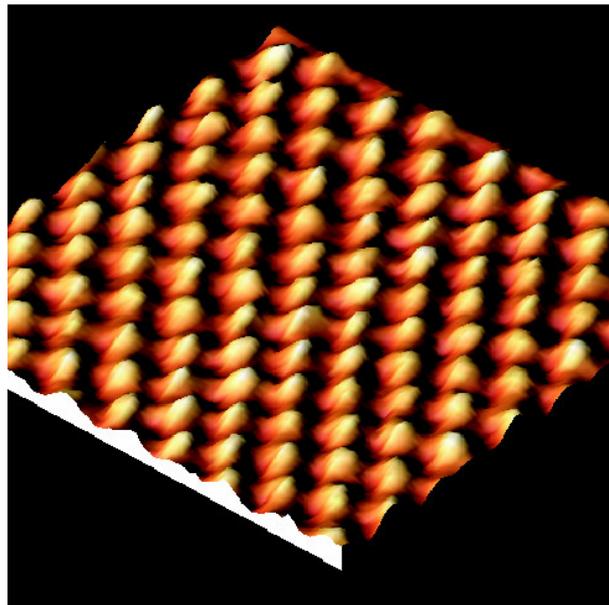


# Scanning tunneling microscopy (STM)

Marco Pratzer and Marcus Liebmann

II. Physikalisches Institut, RWTH Aachen

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## 1 Introduction

Since the first realisation of the scanning tunnelling microscope in 1981 by Binnig and Rohrer [1, 2], a whole range of scanning probe techniques with a wide range of applications has developed in the field of surface physics. All these techniques have in common that the surface of a sample is scanned line by line by a very fine tip and the interaction between tip and sample is measured in a location-dependent manner. With the help of data recording electronics, a map of the interaction  $W(x,y)$  is created. In the device used here, this interaction consists of the quantum mechanical tunnel current between a metallic tip and a conducting sample surface. The term and scanning tunneling microscopy (STM) has become established. The extreme distance dependence of the tunnel current makes it possible to advance the spatial resolution down to atomic dimensions. Even the manipulation of individual atoms has become possible in the meantime. Binnig and Rohrer received the Nobel Prize in 1986 for the development of the scanning tunnelling microscope.

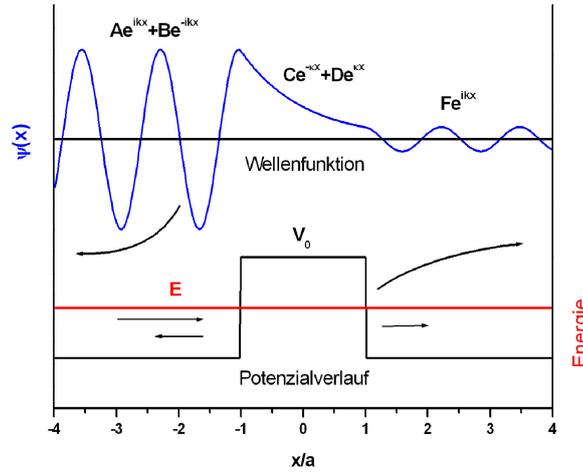
The aim of this experiment is first to get to know the microscope and the determining parameters of the image. This knowledge will then be used to examine an evaporated gold film. Finally, the atomic resolution of the device is demonstrated using a graphite sample (HOPG) and the imaging of the crystal lattice is used to determine the lateral calibration of the scanning range.

These experimental instructions are intended to provide the necessary knowledge for carrying out the experiment. For more information on the theory and applications of scanning tunneling microscopy, please refer to the literature [1, 3–6].

## 2 Theory

### 2.1 Tunnel effect

The basis of the STM is the quantum mechanical tunnel effect, i.e. the fact that under certain circumstances it is possible for electrons to overcome a potential barrier that is greater than their own energy. This would be impossible for a classical particle.



**Figure 1:** Illustration of the tunnel effect. Assume a plane wave approaching a potential barrier from the left. One part is reflected, another part can overcome the barrier.

Let us first consider the one-dimensional case of a plane wave with energy  $E$  incident from the left on a potential barrier of size  $V_0$  and width  $d = 2a$  (Fig. 1) (Note: In reality, the wave function of an electron consists of a wave packet that can be described as a superposition of plane waves with different wavenumbers). It is further assumed that there is a reflected wave and a transmitted wave. For each of the three areas (left, right and in the area of the barrier), the Schrödinger equation can be set up and solved separately. This results in plane waves on the left and right and an exponential course in the middle.

$$\psi_L(x) = Ae^{ikx} + Be^{-ikx} \quad (1)$$

$$\psi_M(x) = Ce^{-\kappa x} + De^{\kappa x} \quad (2)$$

$$\psi_R(x) = Fe^{ikx} \quad (3)$$

Furthermore, continuity conditions for  $\psi(\pm a)$  and the derivative  $\psi'(\pm a)$  at the transition points  $x = \pm a$  have to be applied to the composite wave function. The intensity of the transmitted wave then results from these conditions. The ratio of transmitted to incident current density is called transmission coefficient  $T$  and is calculated in the limiting case of a wide and high barrier ( $\kappa d \gg 1$ ):

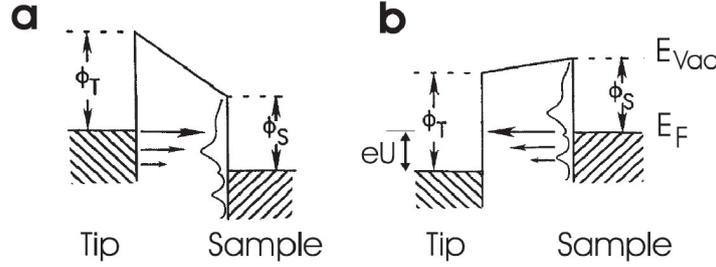
$$T = \frac{16k^2\kappa^2}{(k^2 + \kappa^2)^2} e^{-2\kappa d} \quad (4)$$

with  $k^2 = 2mE/\hbar^2$  and  $\kappa^2 = 2m(V_0 - E)/\hbar^2$ . In our case, the barrier height is determined by the work function of the electrons and is a few electron volts. Typical distances between tip and sample

are in the range of  $10^{-10}$  m. For these tunnel parameters, the transmission coefficient changes by about an order of magnitude if the distance is varied by only 1 Å. This is the reason for the extreme sensitivity of the scanning tunnelling microscope to distance changes.

## 2.2 Tunnel contact in the band model

In a real tunnel contact, not only electrons of a certain energy are involved. Both in the tip and in the sample, the electronic states are filled up to the Fermi energy  $E_F$ . If both sides of the tunnel contact are now at the same electrostatic potential (tunnel voltage  $U = 0$ ), no tunnel current flows because an electron on the other side of the barrier does not find any free states.



**Figure 2:** Energy diagram for the tunnel contact with negative (a) and positive (b) tunnel voltage (from [7]). The electronic states are each filled up to the Fermi energy (shaded area). The barrier is formed by the work functions  $\Phi_T$  and  $\Phi_S$ . An applied voltage  $U$  shifts the Fermi levels against each other by the amount  $eU$ . Electrons from the energy interval  $eU$  are considered for the tunneling process. On the side of the sample, a schematic density of states is also sketched.

Only by applying a voltage  $U$  do the Fermi levels shift against each other and tunneling processes become possible in the energy interval  $eU$ . The tunneling probability decreases exponentially for lower electron energies, since the barrier appears higher for them. Figure 2 illustrates the relationships. How large the tunnel current is, however, depends not only on the barrier height and width, but also on the number of states involved. This is described by the density of states, which indicates how many states are present in a certain energy interval. This does not say anything about whether these states are actually occupied; that is determined by the Fermi energy. The electronic density of states  $\rho(E)$  on both sides of the tunnel contact thus also plays a major role. It can be shown that the tunnel current can be represented as an integral over the applied voltage in addition to the distance dependence:

$$I \propto \int_0^{eU} \rho^S(E_F^S + \epsilon) \rho^P(E_F^P - eU + \epsilon) d\epsilon, \quad (5)$$

where  $\rho^S$  and  $\rho^P$  denote the densities of states of the tip and the sample, respectively, and  $E_F$  denotes the associated Fermi energies, respectively.<sup>1</sup> If one assumes that the densities of states in the energy interval  $eU$  are approximately constant, one arrives at a linear relationship between current and voltage:

$$I \propto U e^{-2\kappa d}. \quad (6)$$

Typical tunnel voltages are in the range of a few millivolts to volts with currents in the order of nA.

If one plots the current as a function of the voltage and the distance, one gains access to two interesting quantities: the density of states and the work function. One can show that the density of states  $\rho(eU)$  is in many cases proportional to the differential conductivity  $dI/dU(U)$ . Experimentally, this quantity is usually measured directly with the aid of a lock-in amplifier. On the other hand, by recording an  $I(z)$  characteristic curve, the exponential part of Eq. 6 can be determined and thus the height of the potential barrier, i.e. the work function. Both the density of states and the work function can thus be determined with spatial resolution using a scanning tunneling microscope.

<sup>1</sup>Strictly speaking, this expression is only valid for  $T = 0$ . At finite temperatures, a corresponding softening of the distribution functions must be taken into account

## 2.3 Piezoelectric effect

In order to exploit the potential of scanning tunneling microscopy in terms of spatial resolution, very precise positioning of the tip relative to the sample in the sub-nanometer range is essential. Any conventional actuator would be hopelessly overtaxed by this. The piezoelectric effect offers a way out here.

Crystals that have at least one polar axis (no inversion symmetry along this axis) can be piezoelectric, i.e. they are able to generate a macroscopic electrical polarization upon mechanical deformation. This is caused by different displacements of different atomic species within the unit cell. If the crystal possesses a permanent electrical polarization even without external pressure, it is also referred to as ferroelectricity. The piezoelectric effect is reversible. A change in the electrical polarization (e.g. by coating the crystal with electrodes and applying a voltage) can therefore also cause a deformation of the crystal. The direction of the deformation depends on the relative orientation of the crystal axes and the electric field. A technically important piezoelectric material is the lead zirconate titanate system (PZT), which achieves length changes in the nm/V range, i.e. in the right order of magnitude for use in a scanning tunneling microscope.

Piezo materials, like ferromagnets, exhibit hysteresis, i.e. the deformation is not only dependent on the applied voltage but also on the previous history. In addition, a piezo reacts to a sudden voltage jump with a rapid deflection, followed by a slow creep. There is also thermal drift, a slow, irregular movement. All these effects can show up as slight image distortions when operating a scanning tunneling microscope.

## 3 Principle of the scanning tunneling microscope

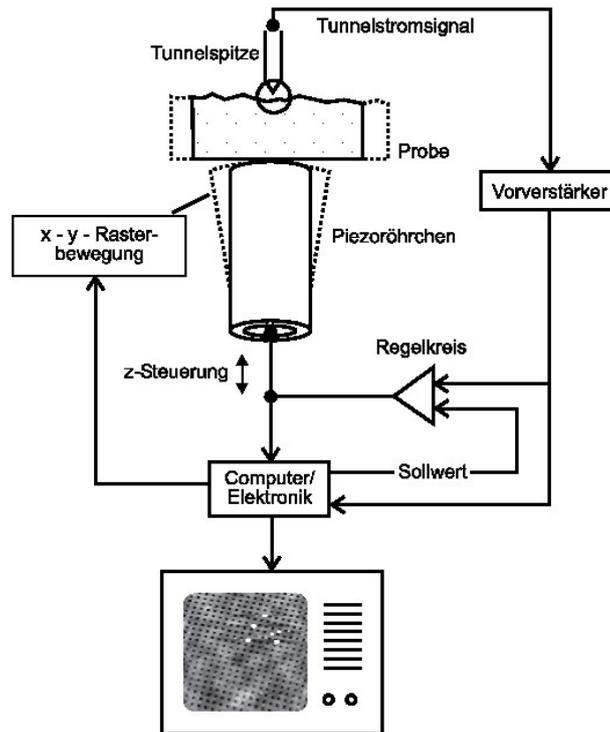
The basic operation of a scanning tunneling microscope is shown in Fig. 3. With the help of one or more piezoelectric crystals, a fine metallic needle, the tip, is scanned over the sample surface to be examined. A tunnel voltage of the order of mV to V is applied between the tip and the sample. The distance-dependent tunnel current flowing between the tip and the sample is measured.

A controller now tries to set the distance between the tip and the sample so that the tunnel current is equal to a setpoint previously set by the user (constant current mode, see below). If one assumes a density of states that does not depend on the location on the sample surface, the tip then follows the topography at a distance that depends on the set tunnel current. As can be seen from Eq. 6, a higher set point of the tunnel current requires a smaller distance between the tip and the sample for the same voltage. Conversely, a larger tunnel voltage for the same current leads to an increase in the distance. In the following, some aspects of the scanning tunnelling microscope will be discussed in more detail.

### 3.1 Realization of the scanning movement

The most common are two different ways to build a piezo scanner to realize the  $x$ ,  $y$ ,  $z$  movement:

- The tube scanner: These are tubes made of piezo material (usually PZT) on which electrodes are vapor-deposited on the inside and outside. If a voltage is applied between the inner and outer electrodes, the tube lengthens or shortens depending on the sign of the voltage. Thus, a distance control ( $z$ -direction) can be realized. Now, one can also divide the outer electrode into four  $90^\circ$  segments and, for example, apply a positive voltage  $+U_x$  to the right segment and a negative voltage  $-U_x$  to the left compared to the earthed inner electrode. Then the tube becomes longer on one side and shorter on the other and thus tilts to the side ( $x$ -direction). With the two remaining segments, one can create a movement in the  $y$ -direction. Typical deflections are a few nm per volt of applied voltage, depending on the geometry of the scanner.



**Figure 3:** Principle of the scanning tunnelling microscope. Control electronics generate the scanning movement with which the sample is moved relative to the tip. The tunnel current  $I$  is measured and compared with an adjustable setpoint in a controller. The control signal  $z$  for the distance control is then calculated from this. Both channels ( $z$  and  $I$ ) can be used for mapping.

- The  $x,y,z$  scanner: Here there is a separate piezo crystal for each spatial direction ( $x$ ,  $y$  and  $z$ ). With three different voltages, the relative position between tip and sample can be controlled. In our case, this type of scanner is present (see also Fig.7) with a control voltage of max.  $\pm 10$  V.

### 3.2 The tunnel tip

To achieve high spatial resolution, the tip with which one scans the sample must be as sharp as possible, ideally with only one atom at the tip. This may seem hopeless at first. However, the extreme distance dependence of the tunnel current comes to one's aid. If one imagines an irregularly shaped tip above a flat sample, it is clear that some atom has the smallest distance to the sample. This atom is then practically the sole carrier of the tunnel current, since the contribution of the second nearest atom is in the vast majority of cases much smaller. A problem occurs when the sample is uneven. Then it is possible that different atoms carry the tunnel current, depending on whether one is on a mountain, valley or slope. In this case, one can obtain multiple images, e.g. of a mountain. Mathematically speaking, the mapping then represents a convolution of the sample geometry with the peak geometry.

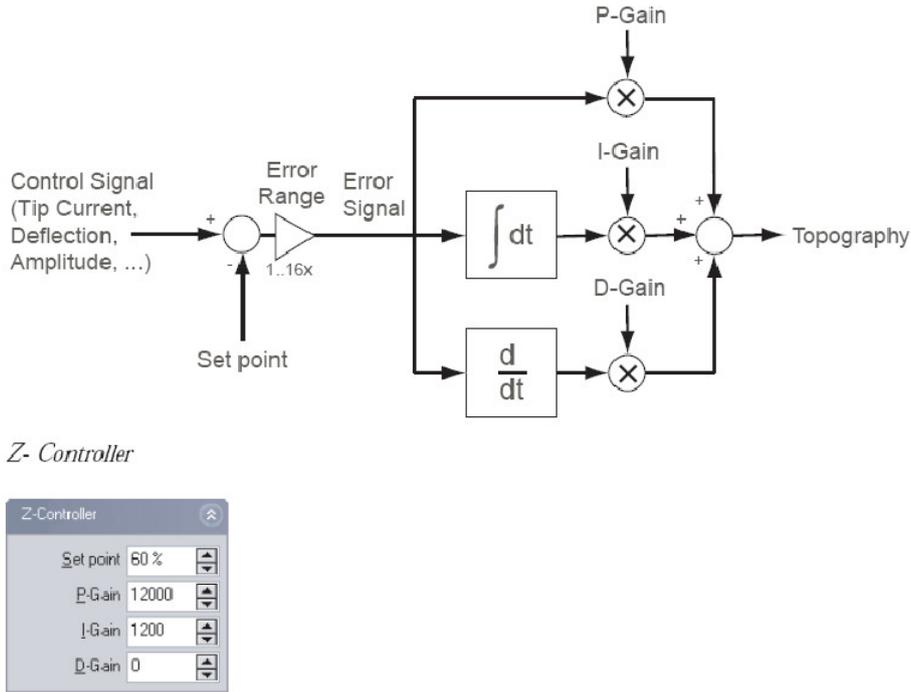
### 3.3 Signal path

Various measurement and control signals are required to image a sample surface. First, the raster voltage is generated in the data acquisition electronics, which produces a lateral relative movement between the tip and the sample. The tunnel current occurring during this movement is converted into a voltage of a few volts via an amplifier circuit and fed to the recording electronics. There, the current signal is compared with the setpoint that was specified by the user before the measurement. This signal can be recorded depending on the grid voltage applied (i.e. the location). A control ensures that if the current is too low (too high), the distance to the sample is reduced (increased).

The  $z$  voltage required for this is passed on to the piezo and also recorded by the electronics. Thus, depending on the setting, one can obtain a map of the measured tunnel current or the  $z$ -voltage.

### 3.4 PID control

In order to generate a useful manipulated variable  $Y(t)$  for controlling the  $z$ -voltage from the measured current signal  $I(t)$  (the controlled variable) and the comparison with the setpoint  $S$ , a suitable algorithm is needed. The most primitive realisation of such a control would be to extend the peak to the maximum when the current is too low and to retract it when it is too high. Of course, this inevitably destroys the peak.



**Figure 4:** Schematic representation of the PID controller in the STM. The corresponding control parameters can be set in the control software menu Z-CONTROLLER.

A better method is to measure the deviation from the setpoint and change the distance proportionally (small deviation, small reaction). This deviation is called the error signal  $E(t) = I(t) - S$ . The associated manipulated variable is then  $Y(t) = gE(t)$ , where  $g$  is a proportionality constant, also called gain in this context. However, this so-called proportional control leads to a permanent control error, as can be seen in the example of a heating system ( $Y$ : heating power,  $I$ : temperature): If the temperature is correct, the heating power is zero, since  $E = 0$ . If the temperature is too low, the heating starts to work; if the temperature is too high, the output is zero. Thus, there can be a constant fluctuation around the setpoint. In the equilibrium case, a temperature is set that is always a little too low for the heating to work at all.

This error can be eliminated by integrating the error signal  $E(t)$  over a certain period of time  $T_I$  and adding it to the manipulated variable:

$$Y(t) = gE(t) + h \frac{1}{T_I} \int_{t-T_I}^t E(\tau) d\tau. \quad (7)$$

As a result, a constant value  $E(t)$  leads to a time-increasing manipulated variable until the setpoint is actually reached. This part is called the integral part. The integration time  $T_I$  can be converted into

a frequency  $BW = 1/T_I$ , which then designates the highest frequency that can still be perceived by the integral part. Faster changes, e.g. oscillations, are integrated out. This frequency is also called the bandwidth of the control.

Now it can happen that the controlled variable changes quickly and must be readjusted accordingly. For this purpose, the tendency of the error signal can be evaluated. A fast variation of the controlled variable then leads to a fast change of the manipulated variable, even before the proportional part intervenes in the control due to a large deviation. This can be achieved by introducing a differential component:

$$Y(t) = gE(t) + h \frac{1}{T_I} \int_{t-T_I}^t E(\tau) d\tau + jT_D \frac{dE(t)}{dt}. \quad (8)$$

The disadvantage, however, is that this also amplifies disturbances from outside and the control loop tries to compensate for them. If, for example, noise occurs in the tunnel current that is caused by the electronics and not by the distance to the sample, the control loop would try to minimise this noise by moving in  $z$ , although the cause lies elsewhere. In addition, overshoot can occur with a high differential setting.

In practice, most electronics for the STM range only work with a  $P$  and  $I$  component. In this experiment, only the integral part in the form of a bandwidth is used.

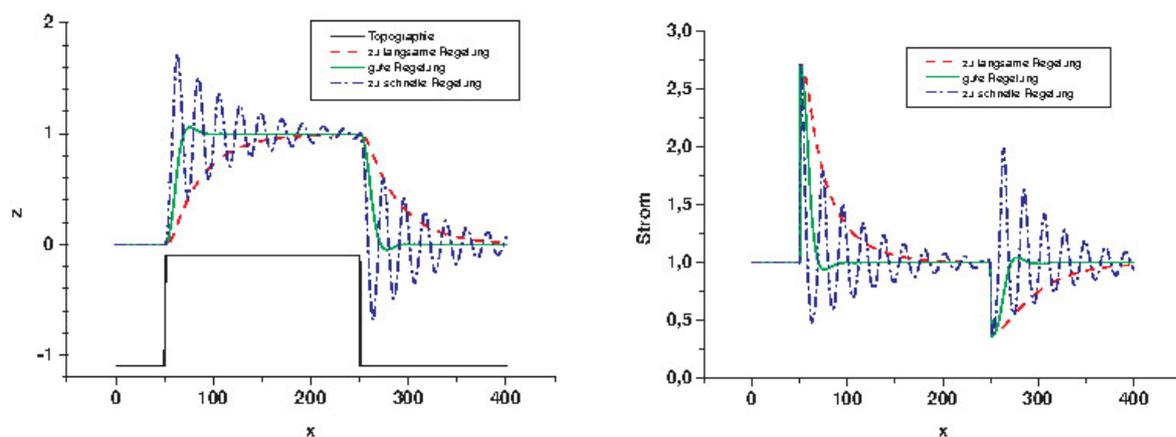
### 3.5 Operating modes and meaning of the measured signals

Depending on the setting in the data acquisition software, one can take an image of the  $z$ -signal or the tunnel current  $I$  during imaging. Both signals are of great importance for the evaluation of an STM image. However, their information content is different for the two typical measurement modes.

#### 3.5.1 Constant current mode

This measurement mode is the standard mode of operation of the scanning tunneling microscope: At a fixed tunnel voltage, a setpoint value of the tunnel current is set, which is achieved by a corresponding control of the tip-to-sample distance ( $z$ ). In this way, the tip can follow surfaces with uneven topography.

The tunnel signal should in principle be constant with a perfect controller, but in practice this is not the case, and conclusions can be drawn from the deviations about the quality of the control and the  $z$ -scan recorded with it. In a way, the current provides an image of the control error.



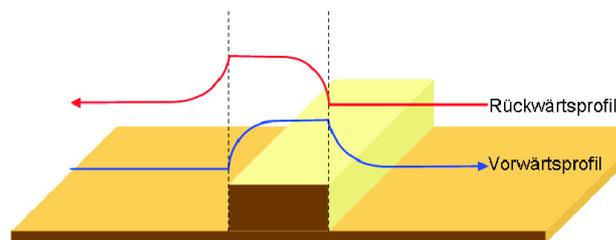
**Figure 5:** Simulation of  $z$  (left) and current (right) signal in constant current mode at different control speeds. The tip moves from left to right. The ideal curve would be a constant distance to the topography (black), well approximated by the green curve. If the controller is too fast (blue), overshoot will occur. If the control is too slow (red), the tip can only follow with a delay.

Figure 5 shows how the STM responds to a sudden change in topography at different control loop settings. The left side shows the true topography as well as the output  $z$  signal, the right side shows the corresponding current signal. The green lines represent a well adjusted controller: after an initial shock, which manifests itself as a maximum in the current signal, the tip reacts quickly with only a small overshoot, and the current settles down again. The overshoot is noticeable in the topography as a slight overshoot of the edge, which also appears to be shifted a bit in the scan direction. While in the  $z$ -signal approximately the topography is represented, one sees in the current signal thus something like the derivative of the topography to  $x$ .

If the feedback is adjusted more sensitively (blue curve), it reacts faster to the edge, but overshoots strongly and settles to the new equilibrium value only after some time. Because of the exponential dependence, this overshoot is usually more noticeable in the current signal and is also first noticed there. In the image, it can become noticeable as a wave pattern in the scan direction. This is then a clear sign that the controller is too sensitive.

In the opposite case of a slow feedback, the current increases strongly at the edge, but the tip reacts only slowly. Accordingly, the current is large over a longer time. You not only get a clear current signal directly at the position of the edge, but also further away from it. This effect is intensified if one slows down the control speed further, so that more and more of the topography becomes visible in the current image. The image impression may appear optically sharper there because of the increased imaging of the edges, but this is not a sign of a good image but of insufficient control. In constant current mode, you want to have the height information in the  $z$ -scan and not in the error signal. In general it can be said that by using a slow feedback you shift more parts of the  $z$ -image into the current image.

The correct feedback parameters in this experiment result as a combination of the values for image size, integration time, tunnel current and scanning time (Sect. 5.3). The larger the image and the faster the tip velocity, the more sensitive the control must be. In this case the  $I$ -value must be increased. However, a too high  $I$ -value can also lead to overshoot.



**Figure 6:** Differences in the forward and backward profile when the feedback is too slow: The island appears shifted in the scan direction. The agreement of both images is a first indication for good control parameters.

Too slow control can be detected quite easily by comparing the images acquired in forward or reverse direction. In case of slow control, the image of islands appears shifted in the scanning direction. If the forward and backward images are the same, this is the first sign that the control speed is correct (Fig. 6).

### 3.5.2 Constant height mode

This measurement method is used especially for atomic resolution on flat surfaces. The tip is moved in a plane parallel to the center sample surface. The feedback is almost off and only compensates for any sample skew. Thus, no significant contrast is expected in the  $z$  signal. However, any unevenness (local distance change) causes a strong variation of the tunnel current, which is not compensated because of the slow feedback loop. One obtains an image of the topography in the current signal. This measurement mode is the limiting case of a too slow controller as described in the last section. The image information has shifted almost completely to the current image.

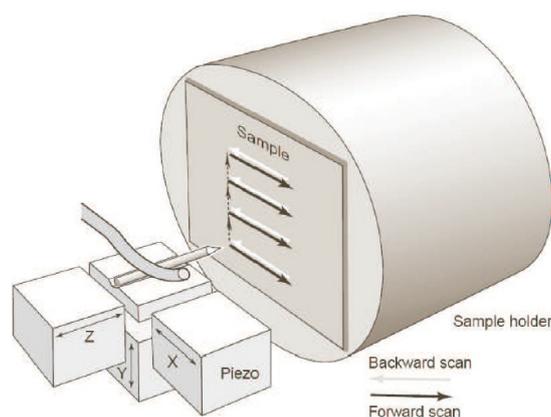
To avoid collision of the tip with the sample in this mode, the sample must be flat on an atomic scale. The average inclination of the surface is compensated with a very slowly adjusted control loop. The advantage is a higher scanning speed and therefore a lower sensitivity to drift and creep effects of the piezo.

### 3.5.3 Overview: Other options for scanning tunneling microscopy

At this point, we will briefly discuss some other interesting possibilities of an STM as used in research today. Since different materials have different electron state densities, one can create a spectroscopy image of the surface by determining the differential conductivity for different tunneling voltages, which can be used to achieve chemical contrast on an atomic scale. For example, if one selects a certain voltage at which a material has a particularly high conductivity, these atoms will appear brighter than the rest in the spectroscopy image.

With the help of magnetic tunneling tips, it is also possible to obtain atomic magnetic resolution. Furthermore, with suitable sample systems, one can also move atoms on the surface and assemble a sample system on an atomic scale.

## 4 Microscope setup

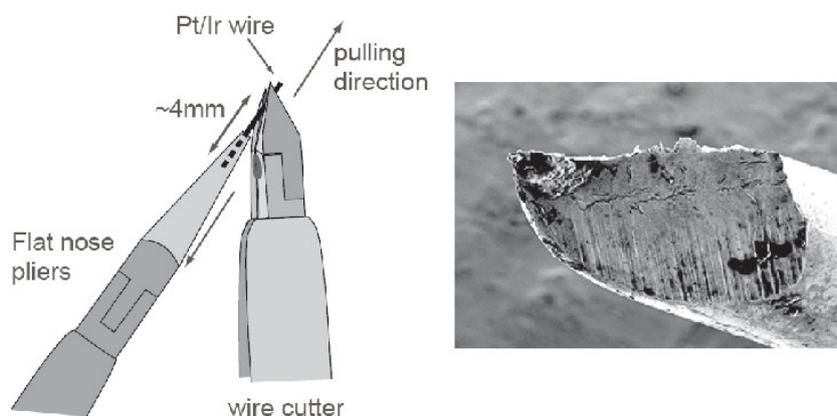


**Figure 7:** Microscope setup. The sample is located on a sample carrier (metal stem), which can be coarse moved with the help of a stepper motor. The tip position is moved across the specimen using the  $x$ ,  $y$ ,  $z$  piezos.

The schematic setup of the microscope used here is shown in Fig. 7. In this type of microscope, during the measurement the sample is at rest and the tip is scanned across the surface using three piezoelectric elements ( $x$ -,  $y$ -  $z$ -direction) to which a maximum voltage of  $\pm 10$  V can be applied. This results in a maximum scan range of 560 nm in the  $x$ -,  $y$ -direction and 200 nm in the  $z$ -direction. The sample is fixed to the sample carrier (metal stamp) by means of a magnetic sample holder. The sample carrier rests on a so-called stepper motor drive, which enables a coarse approach of the tip to the sample. For this purpose, the support of the sample carrier is moved in a sawtooth manner with the aid of another piezo crystal (quickly in one direction and slowly in the other), so that an effective movement of the sample carrier is achieved due to the friction or inertia force. The tunnel voltage is applied between the tip and the specimen and can be varied from -10 V to +10 V. The tunnel current is derived via the specimen, amplified and converted into a voltage (transimpedance converter), which serves as an input signal to the subsequent  $z$  feedback control electronics.

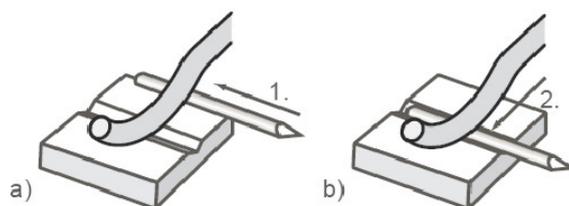
## 5 Operation

### 5.1 Fabrication of the tunnel tip



**Figure 8:** Tip fabrication. Left: Fabrication of a tunnel tip by tearing a wire of platinum-iridium. Right: Scanning electron micrograph of a torn Pt-Ir tip.

In this experiment, the tip is made from a platinum-iridium wire. For this purpose, the wire is cut with a side cutter while twisting and pulling. It is hoped that this will produce a fine tip at the weakest point of the wire at the moment of cutting, where the wire literally flows apart. Experience has shown that simple cutting does not give good results, since in this case only the wire is compressed by the microscopically blunt cutting edges of the tool. Figure 8 also shows an electron microscopic image of a tip produced in this way. At first glance, such a tip appears blunt. However, all that is important for high resolution is that the tunneling current flows essentially across the atom closest to the sample (due to the strong distance dependence of the tunneling current).



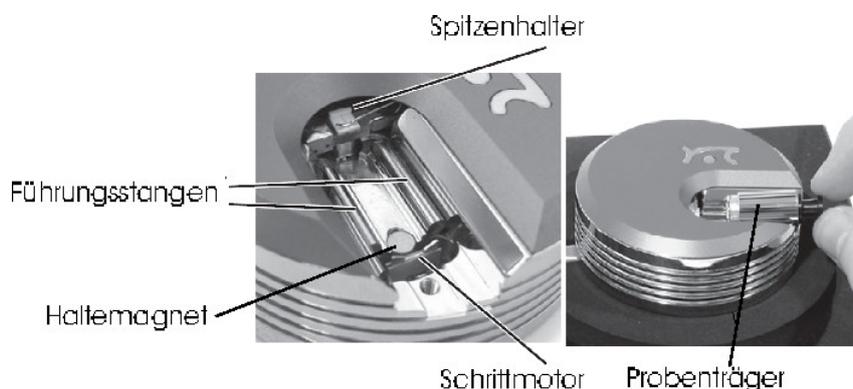
**Figure 9:** Mounting a new tip in the microscope: a) Bring the tip wire parallel to the groove in the tip holder. b) Carefully slide the tip wire sideways under the spring into the groove. If this is too difficult, the spring can be lifted carefully with tweezers.

After the tip has been produced, the wire must still be shortened to about 4 mm and inserted into the tip holder (Fig. 9). Please only lift the retaining spring very carefully with tweezers so as not to damage the scanner. Further tip preparation can be performed in the tunnel contact using voltage pulses. Here, the tunnel voltage is briefly increased so that the high field strength at the tip can throw off individual atom clusters. This in turn influences the imaging properties (although not always positively).

### 5.2 Coarse approach

The approach of the tip to the sample is a particular technical problem of any STM. Since the tunnel current only starts at a distance in the nm range, this distance cannot be controlled optically. Therefore, the approach must be automatic. After setting a tunnel voltage and a set point of the

current, the  $z$  piezo is slowly extended starting from the fully retracted state. During this process, the control loop is active, so it stops the movement when the tunnel contact is reached. If no current is measured even though the piezo is fully extended, it is retracted again and the stepper motor takes a step towards the sample surface. Then the  $z$  piezo is extended again.



**Figure 10:** Coarse approach of the sample to the tip: The sample holder (shown on the right in the picture) is held by the guide rods and rests on the stepper motor drive. The holding magnet serves to increase the contact pressure and thus to amplify the frictional force.

Even before the coarse approach, you should think about appropriate tunnel and feedback parameters. Depending on the starting distance, this approach can take several minutes. Before starting the automatic coarse approach, the tip should therefore be moved by hand with the aid of the stepper motor so close to the specimen that a gap can just be detected. One can take advantage of the fact that most samples are smooth enough to see the reflection of the tip on the sample surface.

**IMPORTANT:** Please note that the sample carrier may only be touched at the handle at the end (do not touch the metal body), as finger grease on the metal body impairs the function of the stepper motor!

### 5.3 Controller software

Microscope control and data acquisition is done with a PC and the EasyScan software. In the left navigator bar, the menus for coarse positioning (“Positioning”), image acquisition (“Imaging”), and measurement parameters (“ $z$  controller”) can be selected. In the following, the most important functions of the software are briefly explained:

#### 5.3.1 Positioning

- Advance: fast approach of sample towards the tip (press and hold key)
- Retract: fast retract of sample from the tip (press and hold key)
- Withdraw: retract sample from tip only a few steps of the motor
- Approach: Sample is automatically approached to the tip and stops as soon as the set tunnel current flows (standard parameter for approach:  $U = 1\text{ V}$ ,  $I = 1\text{ nA}$ ).

#### 5.3.2 Imaging

This window displays the acquired images of the sample: at the top the image of the  $z$  controller (for the forward and reverse scan direction), below it the current image (likewise for both directions) and at the very bottom the latest profiles of the  $z$  signal.

- Image width: width of the scan area (normally, width and height should be equal)

- Time/Line: time for completing one scan line
- Points/Line: number of image points per line (standard: 300 pxl)
- Rotation: scan area can be rotated in  $xy$  plane
- Start: starts scanning (until *STOP* is hit).
- Finish: stops scanning after completing a full image

### 5.3.3 Z Controller

- Setpoint: setting the desired tunnel current
- P: Proportional part of the feedback loop (standard: 1000)
- I: Integral part of the feedback loop (high values = short integration time)
- D: Differential part of the feedback loop (standard: 0)
- Tip voltage: voltage applied to the tip with respect to the sample

## 6 Samples and tasks

### 6.1 Au(111)

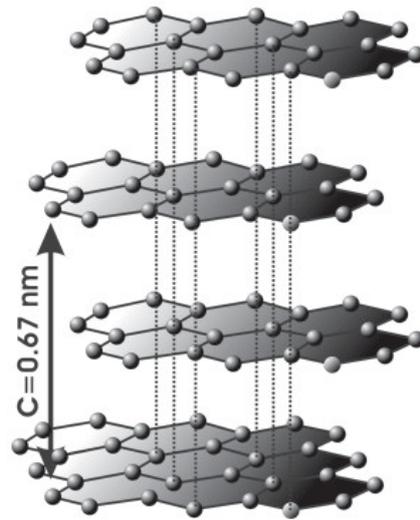
First, the control and imaging properties of the microscope are to be investigated on a relatively uneven sample. For this purpose, a few 100 nm thick gold film is available, which was evaporated onto a glass substrate and then heated. Gold is face-centered cubic (fcc) and forms a (111) surface. The atomic step height is 240 pm.

First, use a large grid area to find a section of the image with easily recognizable structures (e.g. step edges, islands, mountains, etc.). If necessary, zoom into a suitable section of the image. Slightly change the current or voltage (corresponding to the tip distance) to get the sharpest possible image. Attention: in case of very uneven samples, too large a current or too small a voltage will lead to a tip crash. Now vary the integral part of the feedback (4 different settings, the grid time should be approx. 200 ms), and save the images of the  $z$  control signal and the current respectively. In this investigation the interaction with the other parameters like scanning time (time/line), image size and the roughness of the sample is also important.

In general, the measurement is performed in constant current mode. However, if the control velocity is reduced (smaller  $I$  value), the tip can follow the sample surface more and more poorly, which is noticeable in changes in the  $z$  and current image. Increasing the feedback speed can lead to oscillation, which can also be seen in the image. Both extreme cases should be tried and presented in the measurement series described above. What influence does the increase of the integration time have on the reproduction of the topography (e.g. at a gold step edge)? For this purpose, record height profiles of an edge using the profile function of WSxM in the  $z$  image as well as in the current image at the different  $I$  parameters. Make sure that you always place the height profile through the same sample point and determine the slopes. Using the images, the protocol shall discuss the control behavior of the microscope. How does the scanning time affect the image acquisition? Use 4 different scanning times, document the change in the  $z$  and  $I$  image.

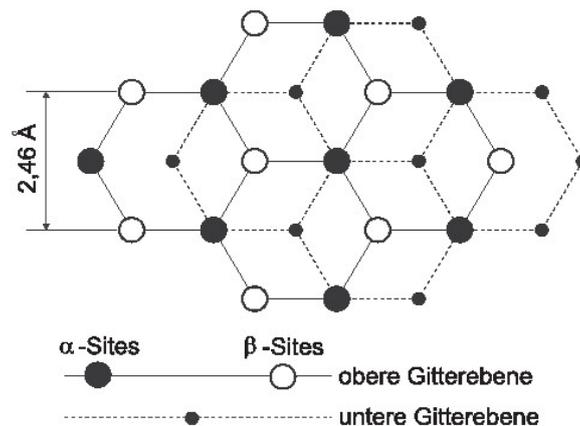
### 6.2 Highly oriented pyrolytic graphite (HOPG)

In the next part, the surface of HOPG shall be imaged to visualize the atomic arrangement. Then, using the known lattice spacing, the calibration for the  $x$  and  $y$  directions will be checked.



**Figure 11:** Stacking sequence of HOPG.

In graphite, the C atoms are  $sp^2$ -hybridized and arranged in layers (Fig. 11), the sequence of which can be characterized by ABAB. In one layer, the atoms form a hexagonal lattice with an edge length of 142 pm. Two layers A and B, whose distance is 335 pm, are shifted among themselves in such a way that only every second atom has a direct neighbor in the layer above or below.



**Figure 12:** Sublattice of HOPG. Atoms labeled  $\alpha$ -site have a direct neighbor below them and are highlighted in the STM image.

For this reason, the atoms of a layer are not equal in their electronic structure, and on the surface every second atom appears highlighted in STM images, so that one often measures a hexagonal lattice with a lattice constant of 246 pm (Fig. 12).

Within a layer, the atoms are linked by localized  $\pi$ -bonds, while the layers are linked to each other only by weak van der Waals forces. The weak forces between the layers manifest themselves in an easy splitting and displacement along the planes.

The respective fourth valence electrons of the C atoms are located in delocalized  $\pi$  orbitals and form a two-dimensional electron gas within a layer, which gives the graphite metallic conductivity parallel to the layers. The conductivity perpendicular to the layers is lower by a factor of  $10^4$  in comparison. Due to this particular electronic and morphological structure, the hexagonal lattice is imaged with a large corrugation in STM images. For this reason, atomic resolution is relatively easy

to achieve on HOPG, and one can therefore use HOPG well to calibrate an STM in the  $xy$  direction.

A HOPG crystal has layers on its surface that can be continuously extended over an area of several  $\mu\text{m}^2$ . This makes it so interesting for scanning tunneling microscopy studies. A piece of adhesive tape can be used to remove the top layers of the graphite single crystal by gently pressing and carefully peeling it away. This results in a clean surface that will not contaminate in air for quite a long time (several months).

First take an overview image of the surface with maximum scan area. The surface should be smooth and have no hills. If the scan lines are all very different, this may be due to an unclean tip. Every now and then you should observe a graphite step edge. Determine the number of atomic layers of graphite step edges in the image using the WSxM profile function. Do the heights match the expected step heights?

In the next step, find a smooth terrace and zoom into it (which scan area is useful to see C atoms?). Optimize voltage and current sensibly until you achieve atomic resolution. Take images in constant-height, as well as constant-current mode (choose feedback parameters accordingly). Are there qualitative differences?

If at least three images with atomic resolution and different grid sizes have been obtained, the calibration of the  $x$  and  $y$  directions of the microscope should be checked and a calibration factor for  $x$  and  $y$  should be determined (calibration factor = (grid size set) / (size determined from image)). Please list in the protocol all images with the measured distances and document each calculation step. Finally, determine the mean value of the calibration factor from the three  $x$  and  $y$  values in each case.

### 6.3 Summary of the task and notes for the protocol

- Gold:

Acquisition of  $z$ -scan and current images for 4 different  $I$ -values of the feedback. Then, the effects of the scanning time (time/line) are to be documented using 4 different values on the image acquisition (i.e., a total of 8  $z$ -scans and 8 current images). How does the topographic image of the surface change? Document the effects on a selected gold structure using profile lines. Find a quantitative measure to evaluate the control, e.g. mean and standard deviation of the current over the whole image or profile lines, squared sum of the difference in the  $z$  image for back and forth scan (per pixel), slope, ...

- HOPG:

Acquisition of an overview image with maximum scan range and at least one graphite step edge. Determination of the step height in atomic layers. Is there a deviation from the expected value? Find a smooth area of the sample surface and zoom into it. Optimize the tip-sample distance using the tunnel current and voltage parameters (how?) so that you get good atomic resolution. Take at least 3 images for different sizes of the grid area in constant height and constant current mode (which feedback parameters are useful for this?). In the protocol, determine the image sizes in  $x$  and  $y$  direction separately (for each image with different image size, i.e.  $3\times$  for  $x$ - and  $3\times$  for  $y$  direction) and compare with the set scan size. Specify an  $x$  and  $y$  correction factor (calibration factor). To what extent does thermal scanner drift or creep play a role? Do not forget the error discussion.

NOTE: Please document each calculation step in the protocol (with error calculation), and always display the measured distances or elevation profiles in the images for traceability.

## 7 Self check questions

- Which physical quantities determine the transmission coefficient of the tunnel process?

- In a real experiment, what is forming the tunnel barrier?
- What do density of states and Fermi energy tell us about the occupation of electronic states?
- Which electrons can contribute to the tunnel current?
- How does increasing the bandwidth of an integral controller affect the imaging?
- How does the image of an edge change when the scan speed is increased?
- What information is obtained from comparing the forward and backward scan directions?
- Why is it possible to go from constant current mode to constant height mode by just changing the integration time of the controller?
- Why must a tunnel voltage of 0 V never be set for image acquisition?
- What should be considered with regard to the tunnel voltage when examining semiconductors?
- How can the image sizes be determined separately by  $x$  and  $y$  directions from an image of the atomic lattice?

## 8 Information on data export

In order for the data to be processed using the WSxM evaluation software, the images must always be exported as a \*.cmv-Datei (XYZ-Points). The import in WSxM is done by the function "heuristic open" (WSxM menu: file -> heuristic open) and parameters: data type: ASCII; Ascii: XYZ; xyz amplitude: m (Meter). Histograms created with WSxM can be saved directly as an ASCII file with the function "save" and read in and edited with QTI-Plot or similar.

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