Chapter 2
Scanning probes microscopes
instrumentation

Objective: learn the general techniques that are essential for SPM.

2.1: Tips

STM tips: requirements

Geometry: Need for atomically-sharp apex for atomic resolution on a “flat” surface, rest of the tip can be blunt.

Strong relief: need for a small angle, otherwise the surface is not faithfully imaged.

The latter point is also true for AFM and related techniques.

An artefact: tip imaging

Sample with needle-like structures: a Al₂O₃ surface imaged by AFM.

The blunt tip is imaged by the sharp sample needle-like structures. Effect recognized by observation of numerous identical structures. Different brightness due to different heights of the needles.
STM tips: materials

Criteria:
Surface quality, low oxydability,
Ease of preparation,
Rigidity.

Materials:
W: easy to etch, rigid.
Pt: very low oxydability, but soft.
Au: very low oxydability but very soft.
Pt$_{90}$Ir$_{10}$: good rigidity/oxydability compromise.

Tip can be simply cut from a wire.
Electrochemistry-based recipies are more reliable.

STM tips: etching techniques

Material-dependent, W is easy.
The reactants flow governs the locality of the etch.

The excess etching time after the drop of the lower part has a strong influence on the final tip geometry:
Long over-etch = blunt tip.

STM tips: final preparation

Used in some specific cases.

Objectives:
Improve tip geometry, remove oxide:
- by thermal treatment: at 1000°C : 2 WO$_3$ + W -> 3 WO$_2$ volatile
- by field evaporation (atom migration)

Attach some atoms of another element (Si, Cu):
- by controlled collision

Chapter 2
Scanning probes microscopes instrumentation

2.2: Basics of piezo-electricity
The piezo-electric effect

Discovered by Pierre and Jacques Curie (1880).

Expansion/contraction $\leftrightarrow$ electrical potential difference

Needs to be an electrically insulating material.

If $z$ and $-z$ are equivalent axes, no piezo-electric response.

Only some anisotropic crystals are piezo-electric: quartz, Rochette salt $KNaC_4H_4O_6-4H_2O$, PZT ceramics …

The PZT materials (1)

Ceramic, solid solutions of PbZrO$_3$ and PbTiO$_3$.

Pb-Zr-Ti = PZT.

Ferro-electric with $T_{\text{Curie}}$ about 250°C, to be used well below.

Polarization process (60 kV/cm, 1h) necessary, aligns dipoles along poling axis ($z$).

The piezo-electric coefficients

Standard notations:

$$
\begin{align*}
\frac{\partial x}{x} & \leftrightarrow 1 \\
\frac{\partial y}{y} & \leftrightarrow 2 \\
\frac{\partial z}{z} & \leftrightarrow 3
\end{align*}
$$

In the linear response regime:

$$
\begin{pmatrix}
\frac{\partial x}{x} \\
\frac{\partial y}{y} \\
\frac{\partial z}{z}
\end{pmatrix} =
\begin{pmatrix}
d_{11} & d_{21} & d_{31} \\
d_{12} & d_{22} & d_{32} \\
d_{13} & d_{23} & d_{33}
\end{pmatrix}
\begin{pmatrix}
E_x \\
E_y \\
E_z
\end{pmatrix}
$$

If voltage applied along the poling axis, only $d_{3...}$ are effectively used:

$${\partial x \over x} = d_{31} E_z \quad \text{most used mode, } d_{31} = 10^{-3} \text{ to } 10 \text{ Å/V}$$

$${\partial z \over z} = d_{33} E_z \quad \text{By symmetry, usually } d_{32} = d_{31}$$

The PZT materials (2)

Piezo-electric coefficients depend strongly on composition and temperature.

$$d_{31} = 1-3 \text{ Å/V}$$

$$d_{33} = 2-6 \text{ Å/V}$$

Fig. 9.3. Definition of piezoelectric coefficients. A rectangular piece of piezoelectric material, with a voltage $V$ applied across its thickness, causes a strain in the $z$ as well as the $z$ directions. A piezoelectric coefficient is defined as the ratio of a component of the strain with respect to a component of the electrical field intensity.
Chapter 2
Scanning probes microscopes instrumentation

2.3: Piezo-electric actuators

The tripod

Used by Binnig et al.
Difficulty to mount, not very rigid.
But easy to operate and to calibrate at the metrological level
(with capacitive sensors).
A bar for every direction:

\[ \delta L = L d_{31} E_z = \frac{L}{e} V \]

Length \( L = 2 \) cm, thickness \( e = 2.5 \) mm, \( V = 250 \) V \((E_{el} = 10^5 \) V/m\), \( d_{31} = 2.5 \) Å/V.
\( \delta L_{\text{max}} = 5000 \) Å, sensitivity = 20 Å/V

The piezo-electric tube

Used by Binnig et al.
Difficult to mount, not very rigid.
But easy to operate and to calibrate at the metrological level
(with capacitive sensors).
A bar for every direction:

\[ \delta L = L d_{31} E_z = \frac{L}{e} V \]

Length \( L = 2 \) cm, thickness \( e = 2.5 \) mm, \( V = 250 \) V \((E_{el} = 10^5 \) V/m\), \( d_{31} = 2.5 \) Å/V.
\( \delta L_{\text{max}} = 5000 \) Å, sensitivity = 20 Å/V

Inner and outer faces are metallized to form two electrodes.
Radial poling, radial electric field : axis 3 = radial direction.
Used to generate a movement in the tube axis direction.

Similar sensitivity but natural rigidity of the tubular geometry.

Example:
Length \( L = 2 \) cm, wall thickness \( e = 0.5 \) mm, \( V_{\text{max}} = 250 \) V \((E_{el} = 5.10^5 \) V/m\), \( d_{31} = 2.5 \) Å/V.
\( \delta L_{\text{max}} = 25000 \) Å = 2.5 \( \mu \)m, sensitivity = 100 Å/V

On drawing, deformation is much exaggerated.
The scanner tube

Tube with again radial poling.
Four quadrant electrodes on the outer face:
$+ V_{x,y}$ and $- V_{x,y}$ applied on two opposite electrodes.
Again, a voltage $V_z$ can be applied on the (single) internal electrode.

$V_Z\ \ V_X\ \ -V_X\ \ V_Y\ \ -V_Y$

The benders

Similar effect as the scanner tube but in a single plate geometry.

Low stiffness, force and resonant frequency.
A scanner built with 4 benders polarized in two halves: no angle with interest for laser beam reflection in AFM.

The shear piezos

The labels 4, 5 and 6 in piezo-electric coefficients describe rotations.

Here, E is applied along direction 1, perpendicular to poling.
The corresponding strain coefficient $d_{15}$ is as high as 11 Å/V.

$L = 1 \text{ mm}, V_{\text{max}} = 250 \text{ V} \ (E_{\text{el}} = 2.5 \times 10^5 \text{ V/m}), d_{15} = 10 \text{ Å/V}$

$\delta X_{\text{max}} = 250 \text{ Å}.$

High forces, used in coarse positioning.
Chapter 2
Scanning probes microscopes instrumentation

2.4: Limitations of piezo-electric actuators

Hysteresis and creep

Hysteresis for various peak voltages.

\[ \Delta L = \Delta L_{t=0.1} \left[ 1 + \gamma \log \left( \frac{t}{0.1} \right) \right] \]

Fragility (it’s a ceramic!)

Mounting rules:
- No pulling force without preload.
- No lateral force or torque.
- Ball tips or flexures to decouple bending forces.
- Ball tips or flexures to decouple lateral forces.
- Bolting of both ends is not recommended.

The scan

Tip is moved to start position, scanned over the surface, and moved back to start position.

Start of the image

Slow scan direction

Fast scan direction

\[ \text{Trace: tip is scanned, topography is recorded.} \]

\[ \text{Retrace: tip is scanned back.} \]
Exemple of hysteresis effect

Difference between trace and retrace images: the image is traced in the hypothesis of a displacement at constant speed, which is not the case.

Closed loop operation

Capacitive position sensor

Closed loop operation: position is measured and corrected. Available in various commercial AFMs.

Other limitations

High voltage amplifiers noise: about 1 mV over 0 to 5 kHz. Induced mechanical noise = about 0.1 Å! (sensitivity 100 Å/V)

Mechanical resonances:

Elongation: \( f_{\text{elongation}} = \frac{c}{4L} \)

Flexion: \( f_{\text{flexion}} = 0.56 \sqrt{\frac{D^2 + d^2}{8}} \frac{c}{4L^2} \)

where \( c = 3 \text{ km/s}, L = 1 \text{ cm}, d = D = 3 \text{ mm} \)

\( f_{\text{elongation}} = 10^5 \text{ Hz}; f_{\text{flexion}} = 10^4 \text{ Hz} \)

Aging: re-calibration needed.

Chapter 2
Scanning probes microscopes instrumentation

2.5: Coarse positioning
how to make approach of a sample towards a tip (from mmm scale down to nm)
The inertial motor

Wagon on two rods, non-sliding condition:
\[ \frac{T}{N} < f \]

\( T = ma \): tangential part
\( N \): normal part of the contact force

If \( a > \frac{fN}{m} \)

the wagon slides.
One step \( \approx \) about 10-50 nm.


The Pan motor

Based on shear piezos.

Walker (inchworm) regime:
Leg by leg movement:
3 stick more than 1.
Retraction of the 4 legs together.

Can also be operated in the inertial regime (4 legs pulsed together).


Example of commercial inertial motors

From Attocube company:

The Pan motor

Based on shear piezos.

Walker (inchworm) regime:
Leg by leg movement:
3 stick more than 1.
Retraction of the 4 legs together.

Can also be operated in the inertial regime (4 legs pulsed together).


Stepper motors

Present in most commercial AFMs, accuracy down to 30 nm.

The electromagnet 1 is turned on, aligning the nearest rotor teeths with the stator teeths. At this point, the rotor are slightly offset from electromagnet 2 teeths. The electromagnet 2 is turned on, ...

Here 25 teeths on rotor, on stator period corresponding to 24 teeths, rotation of 360°/(25x4)=3.6° per cycle.

Works usually only in ambiant conditions (grease needed).
Chapter 2
Scanning probes microscopes
instrumentation

2.6: Design rules and examples

**Vibration isolation**

\[ x_M = \text{difference between tip and base positions: } x_M(t) = x_{M0} \sin(\omega t + \varphi') \]

\[ T_S = \left| \frac{x_{M0}}{x_{S0}} \right| = \sqrt{\frac{\left( \frac{\omega}{\omega_0} \right)^2}{1 - \left( \frac{\omega}{\omega_0} \right)^2} + \frac{\omega^2}{Q^2 \omega_0^2}} \]

\[ T = \left| \frac{x_0}{x_{S0}} \right| = \sqrt{\frac{1 + \left( \frac{\omega}{Q \omega_0} \right)^2}{1 - \left( \frac{\omega}{\omega_0} \right)^2} + \frac{\omega^2}{Q^2 \omega_0^2}} \]

Incoming vibrations / mechanical damping. Practical limitation: \( f_0 > 2 \text{ Hz} \)

**Need for a double stage isolation**

With a 100 nm vibration source at 1 kHz, a \( 10^{-5} \) transfer amplitude gives here a 1 pm vibration on the microscope. \( 10^{-3} \) gives 100 pm = 1 Å.

**With positioning motors**

Very low temperature (60 mK)
AFM-STM with Attocube motors

The beetle geometry

Well adapted to ultra-high vacuum (UHV) systems.

Excellent access to sample and tip.

Coarse approach based on the displacement of the 3 feet along a ramp.


Veeco microscope (at CIME)

Piezo tube for tip scanning
Stepper motors for sample coarse positioning.

STM electronics

Here, the sample is scanned. It could be the tip, no difference.
The tunnel current measurement

\[ V_S = R I_t \]

Bandwidth = \( \frac{1}{RC_{\text{stray}}} = 10^8 \times 10^{-12} = 10^{-4} \) s

Op-amp noise = current noise + voltage noise/wire-ground impedance

\[ i_{\text{OA}} = i_{\text{noise}} + C_{\text{wire}} \omega v_{\text{noise}} \]

about 5 nV/Hz\(^{1/2}\)

Thermal noise of the resistance \( R \):

\[ P_{\text{noise}} = 4k_B T \Delta f \]

\[ i_{\text{Johnson}} = \frac{\sqrt{4k_B T \Delta f}}{R} = 0.3 \text{pA} \]

in \([0;3\text{kHz}]\)

The PID regulation

P, I and D signals generated analogically with op-amps or digitally.

Ziegel-Nichols coefficients setup procedure:

increase \( K_P \) until oscillations appear, reduce \( K_P \) to 0.45 of this critical value, set \( K_I \) to 0.85 the oscillation period. \( K_D \) little useful.


SPM electronics

Here, the sample is scanned. It could be the tip, no difference.

Chapter 3

Imaging with a STM

Objective: to learn the different STM imaging techniques.
Chapter 3
Imaging with a STM

3.1: Imaging principle and techniques

Important parameters

**Voltage bias** determines the energy range of probed electronic states
\[ V_{\text{tip}} - V_{\text{sample}} > 0 : \text{e- from sample to tip, occupied states are imaged.} \]
\[ V_{\text{tip}} - V_{\text{sample}} < 0 : \text{e- from tip to sample, empty states are imaged.} \]

**Scanning frequency**: below the resonance frequency of tubes, below the bandwidth of the tunnel current regulation if active.

**Tunnel resistance** \( R_t \) determines the tip-sample distance. Usually, the current is the parameter:

\[ R_t = \frac{V}{I_t} = 5 \text{ M}\Omega \text{ to } 5 \text{ G}\Omega \]

---

**Constant current imaging**

Tunnel current regulation is active and effective at every time.
\[ I_t = \text{Cst} \]
During the x,y scan, the regulation output \( V_z \) is:
- amplified and sent to the piezo
- measured.

\( V_z(x,y) \) scaled by the Z-piezo sensitivity gives the topography information.

---

**Constant height imaging**

Tunnel current regulation is inactive (or active with a long time constant so that mean slope is followed).
During scan, the regulation output \( V_z \) is close to constant, the tunnel current \( I_t \) is measured.

\( I_t \) gives the topography information.
Possible only on atomically-flat surfaces, highly-sensitive but non-linear information. Much less used.
Chapter 3
Imaging with a STM

3.2: The spatial resolution

The corrugation

Corrugation $\Delta d$ is by definition:

the topography variation amplitude as measured by the microscope.

It is a fraction of Ångström on an atomically flat surface, and can be larger on rougher surfaces.

Depends on every experimental conditions. Can be decreased by a blunt tip, an inefficient regulation ...: not an intrinsic quantity.

Spatial resolution

Binnig (1978); hypothesis of a continuous media.

$$I(\Delta x) = I_0 \exp\left(-\frac{\alpha \Delta x^2}{R}\right)$$ where $\alpha = \frac{\sqrt{2mW}}{h}$

The tunnel current is concentrated on a scale:

$$\sqrt{R} \ll R$$

$R = 10 \text{ nm}$, $\alpha = 1 \text{ Å}^{-1} :$ current flows on a scale $\Delta x = 1 \text{ nm}$.

Spatial resolution is not limited by the tip radius.

Trying to model the atomic resolution

Model of a continuous media surface: homogenous electron gas.

Calculated corrugation $\Delta d_{\text{theo}}$:

$$\frac{\Delta d_{\text{theo}}}{h} = \exp\left(-\pi^2 \left(\frac{R+d}{a^2}\right)\right)$$ (if $a \gg \frac{\pi}{\alpha}$ and $d \gg \frac{2}{\alpha}$)

Stoll (1984)

For a metal: $a = 2.5$ to $3 \text{ Å}$, $k = 1 \text{ Å}^{-1}$, $h = 3 \text{ Å}$. Tip: $R = d = 3 \text{ Å}$

$\Rightarrow$ calculated $\Delta d_{\text{theo}} = 0.01 \text{ Å}$: too small compared to experiment!

This model fails to describe the atomic resolution: the hypothesis of a continuous media is incorrect.
The origin of atomic resolution

Dangling states: anisotropic $d_{z^2}$ localized states close to the Fermi level.

$e^-$ orbitals overlap is strongly modulated during scan.

Contrast depends strongly on the nature of the atoms at the tip apex:
Better resolution on Si after a controlled collision.

Complete description implies including the tip-surface interaction.

Chapter 3
Imaging with a STM

3.3: The STM benchmark Si 7x7

Si (111) 7x7

Si (111) annealed at 1000°C, slow cooling-down.

Single vacancies or adsorbates are visible: true atomic resolution.

Si (111) 7x7 : the model

7x7 reconstruction minimizes nb of pending bonds : 49->19

Miller index refer to the number (= 7) of atomic cells involved.

In STM, the adatoms only are visible, as well as the corner vacancies.

Omicron website: http://www.omicron.de/results
Surface dynamics studies

Real-time dynamics of Pb atoms on Si

Figure 1. Filled and empty state STM images of 0.01 ML Pb on Si(111) 7×7 measured at room temperature. The scanning area is 16.25 × 16.25 nm² [(a) and (b)] and 6.0 × 6.0 nm² [(c) and (d)]. Sample voltages are +2 V [(a) and (c)] and −2 V [(b) and (d)]. Tunnel current is 0.3 nA for all images.

Figure 2. Successive frames e–m measured at 78 K, showing Pb and the formation of a pair [e] of dimers. The scanning area is 14.75 × 14.75 nm². Sample voltages are −2 V. Tunnel current 0.2 nA, measured from 78 to 85 K.