Lecture 2: Measurements with contact in heat transfer: principles, implementation and pitfalls
PART 2

Thermal measurements using Scanning Thermal Microscopy (SThM)

Micro and Nanoscale measurements

Séverine Gomès
• The technique
  • Principles
  • Probes
  • Operating modes
• Approaches of the measurement
  • Energetic balance
  • Modelling
  • Calibration
• Main current issues
Scanning thermal microscopy: SThM

The technique

- General principles
- Cantilever and probes
- Operating modes
SThM methods
operate by:

- bringing a sharp temperature sensing tip in close proximity to the solid surface to be characterized.

- using the variation of the tip temperature depending on the changes of the heat transfer between the tip and sample surface.

\[
\text{SThM} = \text{Scanning Probe Microscopy technique} + \text{Small probe used as thermal sensor}
\]
AFM-based systems

- Observation of all kinds of materials

- Enable to obtain:
  - Force-distance curves
  - Surface topography with the control of sample-probe contact force
General principles

AFM-based systems

- Observation of all kind of materials

- Enable to obtain:
  - Force-distance curves
  - Surface topography with the control of sample-probe contact force

  and independently:

  - Thermal signal versus probe – sample force & distance
  - Thermal imaging and measurement according to the principle of thermal probe
Since 1993, various thermal methods based on the use of different thermosensitive sensors or phenomena

Classification according to the temperature-dependent mechanism used:

- **Thermovoltage**: - Tunneling thermometry and point contact method
  - Built-in thermal sensors: Thermocouple
  - Schottky diode

- **Change in electrical resistance**: Built-in thermal sensors

- **Fluorescence**: Fluorescent nanocrystal glued at the end of a AFM tip

- **Thermal expansion**: - Bilame effect
  - Measurement of the dilatation of the sample surface induced by its internal or external heating
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→ focus on thermovoltage-based and resistive methods.
Cantilevers and thermocouple probes

Some examples

Thermoelectrical voltage = g(T)

Lateral spatial resolution < 50nm

[Shi et al. 2001, J Microelectromechanical Sys 10, 370]

The miniaturization of the cantilever, the tip and the junction at the tip end could lead to a decrease of the probe thermal time constants and to an improvement of the spatial resolution.


Au–Cr thermocouple SThM probe.
Electrical resistance = \( g(T) \)

Wollaston cantilever with a resistive Pt/Rh\(_{10}\%\) wire probe
- Section: 5 \( \mu \)m
- Length: 200 \( \mu \)m
- V shape - curvature radius: 15-20 \( \mu \)m
- Spatial resolution: micrometric

Thin film Pd resistor located at the tip of a contact mode AFM probe
- Cantilever made of Silicon Nitride (Si\(_3\)N\(_4\)).
- Tip radius lower than 100 nm.
- Spatial resolution < 80 nm

[Kelvin Nanotechnology, Glasgow Univ.]
All resistive metallic probes can be used in **two modes**

### Passive Mode
for thermometry

- **very small current through the probe**
- ⇒ **minimal Joule self-heating and enables the measurement of the electrical resistance.**

During a scan, heat flows from the hot sample to the probe and changes the electrical resistance $R_p$ of the probe

### Active Mode
for thermophysical properties analysis

- **larger electrical current through the probe**
- ⇒ **significant Joule heating of probe**

**Probe = heat source + thermal sensor**

Part of the Joule power flows into the sample, depending on its thermal properties. The probe temperature is monitored by measuring the probe voltage. This temperature can be related to the thermal conductivity of the sample.

Under both modes, dc, ac or both measurements

**ac regime** ⇒ **improved signal-to-noise ratio, since lock-in detection is possible.**
Thermal contrast imaging & measurement

Spatial resolution depends on three main factors:

- Tip sharpness
- Thermal design of the probe
- Tip-sample heat transfer mechanisms (surrounding environment)

Temperature field (sample surface)
when the tip comes in local equilibrium with the sample

Thermal conductivity contrast
if the temperature change is determined for a known heat flux

SThM methods
Enable:
- Thermal contrast imaging & measurement

Temperature map of a MESFET (metal/semiconductor field-effect transistor)

SThM image obtained at the level of the interface between irradiated and non-irradiated areas of meso-PSi (85% porosity).

Scanning thermal microscopy: SThM

SThM measurement approaches

Thermometry in steady-state regime and passive mode
Thermophysical measurement (active mode)

- Energy balance
- Modelling
- Calibration
As in all thermal measurement method involving a sensor in contact or in proximity with the sample surface to be characterized, the **Analysis of the energy balance of the system:**

**probe - sample –surrounding environment**

**required**

Probe used in passive mode and in contact with a hot sample.
Energy balance

Thermometry in steady-state regime and passive mode

Thermal resistance network model

As at larger scale, $Q_{s \rightarrow t}$ generates an error!
Thermal resistance network model

In the simple case of a sensitive element at the tip apex, which is the case for almost all the thermovoltage-based SThM probes \( T_{t,c} = T_{\text{probe}} \), and no heat transfer between the cantilever and the sample surface,

\[
Q_{s-t} = \frac{T_s - T_{\text{probe}}}{r_{th,s} + r_{th,c}} = \frac{(T_{\text{probe}} - T_a)(r_{th,pe} + r_{th,cant.})}{r_{th,pe} r_{th,cant.}}
\]

Expression of the correction to be applied to the nominal measurement of the instrument \( T_{\text{probe}} \):

\[
\delta T = T_s - T_{\text{probe}} = \frac{(T_{\text{probe}} - T_a)(r_{th,s} + r_{th,c})}{r_{th,pe}}
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Energy balance

Thermometry in steady-state regime and passive mode

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Expression of the correction to be applied to the nominal measurement of the instrument $T_{probe}$:

$$\delta T = T_s - T_{probe} = \frac{(T_{probe} - T_a)(r_{th,s} + r_{th,c})}{r_{th,p}}$$

If the error $\delta T$ is unknown, only the probe tempertaure can be measured.
Thermal resistance network model

- Heating of the sample by the probe operating in active mode (in dc or ac regimes)

\[ Q_{t,s} \text{ written as a function of:} \]
- the thermal power \( P \) used for the heating of the probe,
- the measured probe temperature \( T_{\text{probe}} \)
- the thermal property to be determined

\[ Q_{t-s} = \frac{(T_{t,c} - T_a)}{(R_{th,s} + R_{th,c})} = P \]

\[ - \left( \frac{(T_{t,c} - T_a)}{R_{th,\text{env}}} + \frac{(T_{\text{probe}} - T_a)}{R_{th,p}} \right) \]
**Energy balance**

**Thermophysical measurement (active mode)**

- **Heating of the sample by the probe operating in active mode (in dc or ac regimes)**

  $Q_{t,s}$ written as a function of:
  - the thermal power $P$ used for the heating of the probe,
  - the measured probe temperature $T_{probe}$
  - the thermal property to be determined

  \[
  Q_{t-s} = \frac{(T_{t,c} - T_a)}{(R_{th,s} + R_{th,c})} = P - \frac{(T_{t,c} - T_a)}{R_{th,env}} + \frac{(T_{probe} - T_a)}{R_{th,p}}
  \]

- **For bulk and homogeneous thick samples,**
  the **sample thermal conductivity** $\lambda_s$ included in the expression of the sample thermal spreading resistance $r_{th,s}$

  \[
  r_{th,s} = \frac{1}{4\lambda_s b}
  \]

  *Heated area on the sample surface assumed circular of effective radius $b$ and isothermal.*

The ideal situation for an easy estimation of the sample thermal conductivity should be that

- $\lambda_s$ is included in the expression of $r_{th,s}$ only,
- the tip apex temperature $T_{t,c} = T_{probe}$.

In this case:

$$Q_{t-s} = 4\lambda_s b(T_{t,c} - T_a) / (1 + 4\lambda_s b r_{th,c})$$

For $Q_s$ to depend primarily on $\lambda_s$, $4b\lambda_s r_{th,c} << 1$ must be satisfied:

the dominant thermal resistance should be that of the sample.

→ SThM measurements mainly sensitive to $\lambda_s$ for materials of low thermal conductivity.

Energy balance

Thermophysical measurement (active mode)

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$\rightarrow$ SThM measurements mainly sensitive to $\lambda_s$ for materials of low thermal conductivity.

However,
- $T_{t,c} \neq T_{\text{probe}}$
- resistive probes have an extended thermosensitive element.

Calibration curve

$G = 1/r_{th,c}$

$G = f(k_s)$

$G = k_{\text{bulk}} / k_s$
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$r_{th,c} : 5 \times 10^{-6} \text{ K.W}^{-1}$ to $8 \times 10^{-6} \text{ K.W}^{-1}$
b : several micrometers

However,
- $T_{t,c} \neq T_{probe}$
- $r_{th,c}$ and $b$ strongly depend on $\lambda_s$

$Q_{s-t}$ or $Q_{t-s}$ and then the error $\delta T$ (in thermometry) depend on many parameters characterizing:

- the surrounding gas (pressure, temperature, degree of relative humidity),
- the tip-sample mechanical contact: mechanical properties of tip and surface, tip-sample force, surface roughness and topography,
- the thermophysical properties of probe and sample.

- The estimation of all the involved parameters through modelling is not trivial and is still one of the main limitations of SThM involving nanoprobes

- Experimental calibration methodologies proposed
Measurement modelling

Depending on the probe, various analytical and numerical models proposed to link the nominal signal effectively measured and the parameters to be determined.

The most used analytical thermal model based on the resolution of the heat equation in the probe described as thermal fins.

Models consider
- geometrical and dimensional parameters
- the physical properties of materials

Models used to determine:
- **unknown modeling parameters** from comparison with measurements in well controlled configurations (surrounding, reference samples, …) through calibration

Models consider:
- effective parameters such as:
  - an effective coefficient $h$: heat losses by the whole probe surface to its environment,
  - an effective thermal resistance $r_{th,c}$
  - an effective thermal radius $b$.

- an appropriate description of the sample depending on its shape, …
Example:

\[
\frac{\partial^2 \theta}{\partial x^2} - \frac{hp}{\lambda S} \theta + \frac{\rho I^2}{\lambda S^2} (1 + \alpha \theta) = \frac{1}{a} \frac{\partial \theta}{\partial t}
\]

\[
\lambda S \frac{\partial \theta}{\partial x} \bigg|_{x = \frac{L}{2}} = G_{eq} \theta(x = \frac{L}{2})
\]

\[
1/G_{eq} = R_{th,c} + R_{th,s}
\]

Sample thermal resistance

Contact thermal resistance
• The fitting of simulated measurement with experimental data in well-known conditions (various surrounding conditions, various frequencies of heating of the probe, various samples...) → identification of the modelling parameters $h$, $R_{th,c}$, $b$ and the probe characteristics such as its time response.

Measurements under ambient conditions, no contact with a sample

low pass filter of cut-off frequency about $f_c = 800$ Hz. Once $f_c$ is experimentally determined, the effective radius $r_p$ and the effective half-length $L_p$ of the tip can be determined from the modeling.

• Once calibrated for a given experimental configuration (probe and surrounding environment), the modelling is used to characterize unknown specimens from measurements performed in the same configuration.
Calibration methods implemented for the determination of $\delta T_p$

- Use of laboratory self-heating samples.

   Self-heating samples fabricated for SThM calibration include:
   - instruments that are specifically designed for absolute temperature measurements on the scale of one micron: Johnson noise in a small metallic resistor, or instrumented membrane.
   - hot sources implemented in subsurface volume with a metallic line heated through joule effect.
     - Samples
       - generally heated in ac regime to demonstrate thermal mapping with low signal-to-noise ratio.
       - used to characterize the dynamic response of sensors, which is also an important parameter to be considered.

- Based on comparisons of SThM measurements with:
  - either measurements obtained by optical thermometry methods,
  - either results of simulation of the sample surface temperature (or both).
Calibration methods implemented for the determination of $\delta T_p$

However,

- Optical thermometry methods have a spatial resolution limited to few hundred nanometers
- Simulations at micro and nanometric scales are often dependent on simplifications or critical parameters.

These comparisons are then not strictly applicable to SThM temperature measurements with a spatial resolution of a few tens of nanometers.

Accurate temperature measurements after calibration are only possible on samples that hold almost the same surface properties than the ones of the calibration samples.
Other example of calibration (purely experimental)

The calibration can be performed with a set of experiments involving flat bulk samples of well-known thermal conductivities in a range that covers the expected value of the thermal conductivity $k_s$ to be measured.

Practically, the tip heated with an increase of temperature larger than 80 K to ensure a good signal to noise ratio and avoid issues related to the presence of a water meniscus.
Current issues

What is about the smallest probes?
Sensitivity to thermal conductivity reduced compared to the one of larger and metallic probes due to nanoscale size effects.

$L_{ph}$ in pure crystalline Si $\approx 300$nm (Ta) $\gg$ sizes of the tip apex of the Si nanoprobe.

Macroscopic description does not accurately model the heat flow and the heat conduction through the tip is modelled as quasi-ballistic transport.

- Additional thermal resistance to describe heat transfer at the probe apex.
- Nanoscale size effects on the sample spreading resistance.
A better understanding of the thermal interaction between a SThM tip and a sample is crucial for a good understanding of measurement and a good interpretation of the contrast of thermal images.
Various mechanisms of heat transfer

Various length scale to be considered

- Ballistic conduction through air
- Diffusive conduction through air
- Conduction through water meniscus
- Conduction through solid-solid interface

Near field heat transfer

Far field heat transfer

Heat from tip to sample

$d < \Lambda_{\text{air}}$

Tip apex

Sample surface

Tip

Sample

$T_{\text{tip}} > T_{\text{sample}}$

$\lambda_{\text{rad,th, max}}$

$\lambda_{\text{rad,th, max}}$

Atomic scale and roughness

Probesample heat transfer
Complex

- Various mechanisms of heat transfer
- Various length scale to be considered

**Probe-sample heat transfer**

- **Heat from tip to sample**
  - $d > \Lambda_{\text{air}}$
  - Tip apex
  - Sample surface

- **Conduction through water meniscus**
  - Tip apex
  - Sample surface

- **Conduction through solid-solid interface**
  - Atomic scale and roughness

- **Diffusive conduction through air**
  - Tip
  - Sample

- **Ballistic conduction through air**
  - Probe roughness

**Near field heat transfer**

**Far field heat transfer**

General neglected if not specifically studied
If not under vacuum conditions, a large area of the probe is in contact with the air lying around and can heat it. Hot air can then heat the sample. The hot part of the tip can extend over tens of micrometres in some SThM probes → the heated part of the sample could potentially be as large: 

heat transfer mechanism extremely efficient. 

→ impact on spatial resolution of SThM.

Thermal imaging of Silicon nanowires (diameter lower than 100nm) in SiO2 matrix
Different heat transfer mechanisms depending on the probe-sample distance

*When the heater is far from the tip end and therefore from the sample*, strong probe-sample air heat transfer also occurs, with air being heated at the area of the conical tip or directly by the cantilever.

*When the tip-sample distance is large*, heat convection can occur: the heat flux will be carried by the motion of air generated by the temperature difference

→ *A large part of the heat can be lost in the air* because heat lost by a hot tip does not necessarily flow towards the sample.

*At shorter distances (few micrometres and below)*, 
heat diffusion: main mechanism of heat transfer through the air. 
The heat transfer can then be modelled with standard Finite-Element Modelling (FEM) tools.
Different heat transfer mechanisms depending on the probe-sample distance

*When the distance reaches few hundreds of nanometres, ballistic heat transfer:*

The mean free path of the heat carriers $L_{\text{gas}}$, here air molecules (mostly nitrogen ones), becomes comparable with the tip-sample distance ($L_{\text{air}} \sim 70\text{nm at } P=1\text{ bar}$)

When $d < L_{\text{gas}} \left( Kn_{\text{gas}} >1 \right)$, the continuum assumption is not appropriate. This concerns the very end of the tip where $d$ is smaller than some µm, or when the probe is operated in very low vacuum ($Pressure <1 \text{ mBar}$).
Reminder

$G_{\text{air}}$ from $10^{-8}\text{W.K}^{-1}$ to $10^{-6}\text{W.K}^{-1}$ depending on the size of the tip.

$G_{\text{total}}$ as a function of the probe mean temperature $T_m$

$T_a = 30^\circ C, RH = 40\%$.

Good agreement with previous works of Majumdar et al. with a SThM thermocouple probe with nanometric curvature radius

$G_{\text{total}} = 6.7\text{ nW/K}$
The radius of the mechanical contact $a$ generally estimated from mechanical contact theories.

These theories apply to the contact between a sphere and a flat surface and their use leads to consider an ideal contact between surfaces of perfect quality.

Whatever the considered SThM tip, $a$ estimated to be lower than a few tens of nm.

For such nanoscale contacts, phonon mismatch and mechanical contact geometry to be accounted.
→ Thermal boundary resistance to be considered due to the difference in phonon dispersion between the two materials in contact.

\[ R_{th,\text{contactB}} = \frac{R_{th,B}}{\pi b_{c,p}^2} \]

\( R_{th,B} \) a thermal boundary resistance that has the same units as the bulk thermal contact resistance.

The values of \( R_{th,B} \) experimentally determined for solid-solid contacts near room temperature typically lie in the range \( 5 \times 10^{-7} - 5 \times 10^{-9} \text{ m}^2\text{.K.W}^{-1} \)
However in reality

The tip-sample interface never perfect:

- contamination or oxide layers can cover surfaces
- contact area is non-continuous due to weak coupling bonds between the atoms of solids or surface roughness
Heat transfer through solid-solid contact

- Due to the roughness of the tip and sample surfaces, the apparent contact surface is not continuous.
- The surface can be divided into smaller contact area.
- Appropriate modelling of this effect depends on the ratio of the averaged mean free path of heat carriers $\Lambda_c$ to the size $b_c$.

If $b_c >> \Lambda_c$, diffusive transport applies. Classical solutions are applicable:

$$G_{th,contact} = K k_s b_c$$

where $K$ is a geometrical factor describing the heat spreading within the sample.

If $b_c < \Lambda_c$, ballistic solutions must be considered.

When $b_c <<\Lambda_c$, the notion of finite contact spots may be extrapolated to the atomic scale.

Quantization of thermal conductance?
Conclusions
Accurate temperature measurement is not an easy task whatever the considered length scale is.

Errors depend on:
- thermosensitive phenomena,
- the sensor which can create local temperature disturbance and therefore bias.

Very often, this latter error is ignored.

In this lecture dedicated to contact thermal measurement, one have tried to provide to the readers the know-how in various situations (temperature measurement in fluids or opaque medium) in order to perform the best measurements as possible.

Thermal measurement at micro and nanoscales using Scanning Thermal Microscopy were presented.

The crucial step of calibration, the limitations and current issues of the method were given.

The complexity of the tip-sample heat transfer at micro and nanoscale was discussed. This explains why its deep understanding is required in order to obtain the best achievements of the techniques.
Thanks a lot for your attention!

Questions?