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Measurement system for thermophysical properties of thin films in a broad temperature range

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Outline

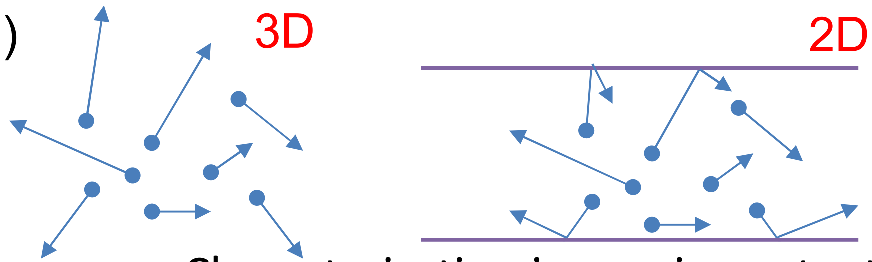
1. Motivation & Project Goals
2. Concept
3. Sensor layout
    1. El. conductivity measurement
    2. Hall constant measurement
    3. Seebeck coefficient measurement
    4. Thermal conductivity measurement
4. Measurement setup
5. Applications
6. Summary

Motivation

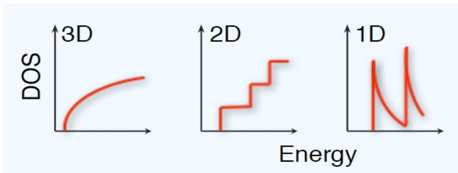
- Physical properties of thin films differ from bulk material
    - Parasitic surface effects are much stronger due to smaller dimensions and high aspect ratios

➡ e.g. Boundary scattering & Quantum confinement

a)



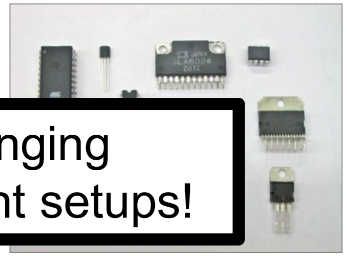
b)



- Characterization is very important for both, research and technical applications

Thermal barrier coatings

Thermoelectric devices



Integrated devices

Characterization of thin film samples can be challenging
➡ growing demand for thin film measurement setups!

Project goals

- Development of a high quality, easy to use characterization system for thin films
- Temperature dependent measurements with easy sample preparation and handling
- High measurement flexibility (material, thickness, deposition methods)
- Consistent results -> **all measurements are taken in the same direction (in-plane)**
- All measurements should be done at only one sample within one run
    - To avoid errors due to:
        - different sample compositions
        - different sample geometries (e.g. size or thickness)
        - different heat profiles
        - different environmental conditions

➡ Measured properties are comparable to each other

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- Measured parameters:

→ $$ZT = \frac{S^2\sigma}{\lambda}T$$ // Figure of merit

+ A_H // Hall Constant

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$$\Rightarrow \quad ZT = \frac{S^2 \sigma}{\lambda} T$$

// Seebeck Coefficient

$$+ \quad A_H$$

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- Measured parameters:

→ $ZT = \frac{S^2\sigma}{\lambda}T$ // electrical conductivity

+ A_H

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- Measured parameters:

$$\Rightarrow ZT = \frac{S^2\sigma}{\lambda}T \; + \; c_P \; + \; \varepsilon$$

$$+ \; A_H$$

// thermal conductivity

+ specific heat + emissivity (depends on sample)

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- All measurements should be done at only one sample within one run
- Measured parameters:

$$\Rightarrow \quad ZT = \frac{S^2\sigma}{\lambda}T$$

$$+ \quad A_H \Rightarrow \mu + n$$

// Hall constant (➡ calculation of charge carrier concentration and mobility)

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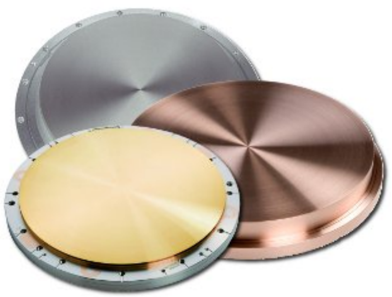
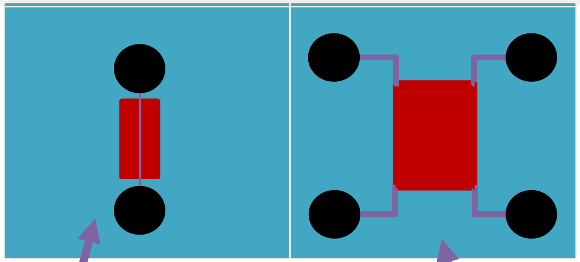
Concept



Sample material

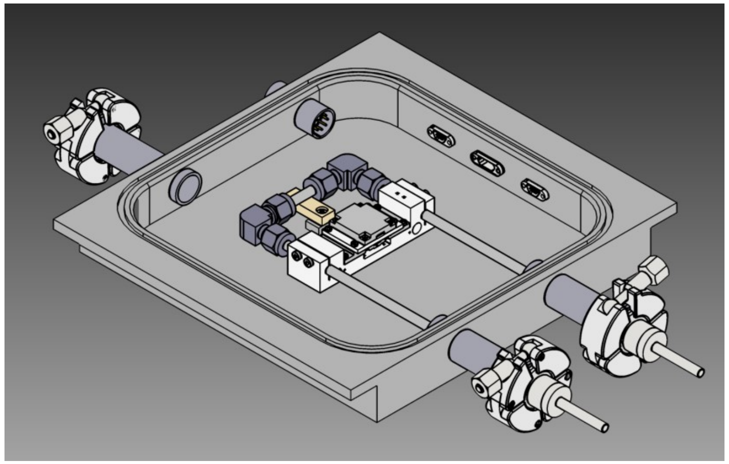
Sample deposition

sputtering, evaporation, ALD, ink-jet printing, spin coating etc.

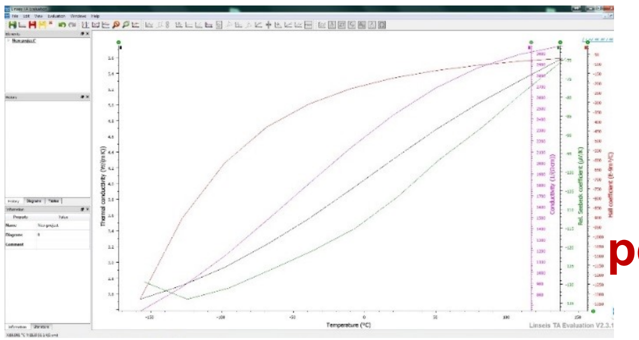


Pre-structured measurement sensor

Sample connection



perform measurement



Measurement and evaluation software

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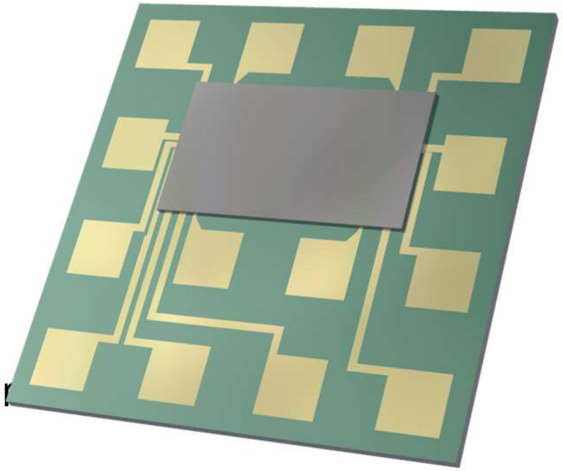
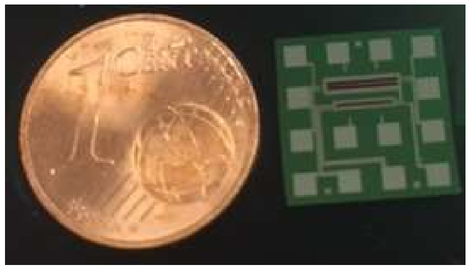
Measurement Sensor

- Chips are completely pre-structured and ready to use.
- Based on Si-substrate.
- Lithographically defined structures (cleanroom process)
  - Pt electrodes for connecting the sample.
  - Membranes for thermal conductivity measurements.
  - Resistance thermometers and "on the chip" heater for Seebeck coefficient measurement.
  - Surface for deposition is Al_2O_3.
- Deposition mask for well-defined sample geometry (strip off foil mask or metal shadow mask).
- Sample deposition on ONE side in ONE process



CAD drawing of measurement chip.



Size of the chip

➡ Goal: Easy to use chips with a broad application range and a minimum effort for the sample preparation.

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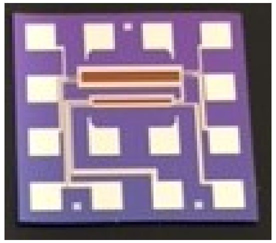
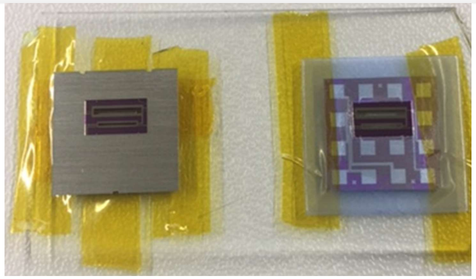
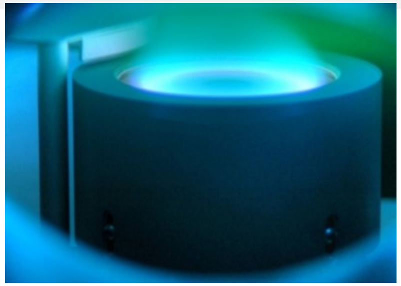
Sample preparation route (shadow mask)



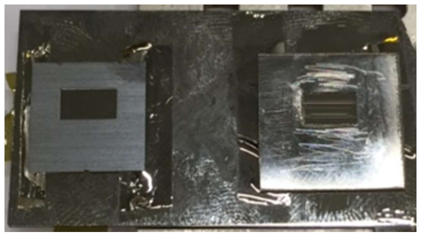
Empty sensor
without mask



Sensor with attached shadow masks
(metallic mask left, strip off mask right)

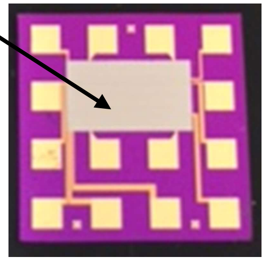


Sample deposition
(e.g. magnetron sputtering)



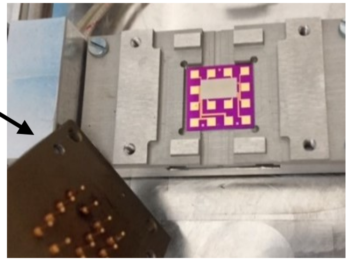
Sensor covered with sample

Sample



Sensor with defined
sample on top

Lid with
spring pins



Insert sensor into sample holder
and perform measurement

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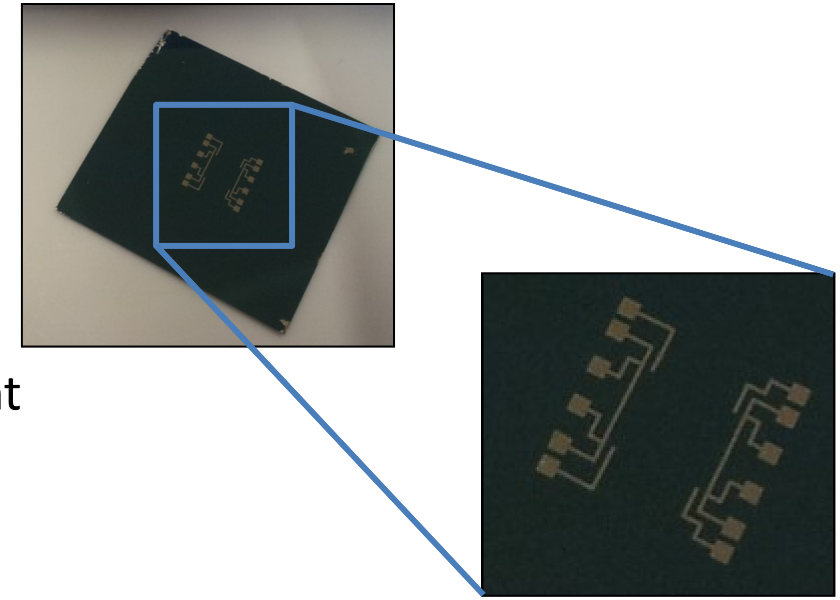


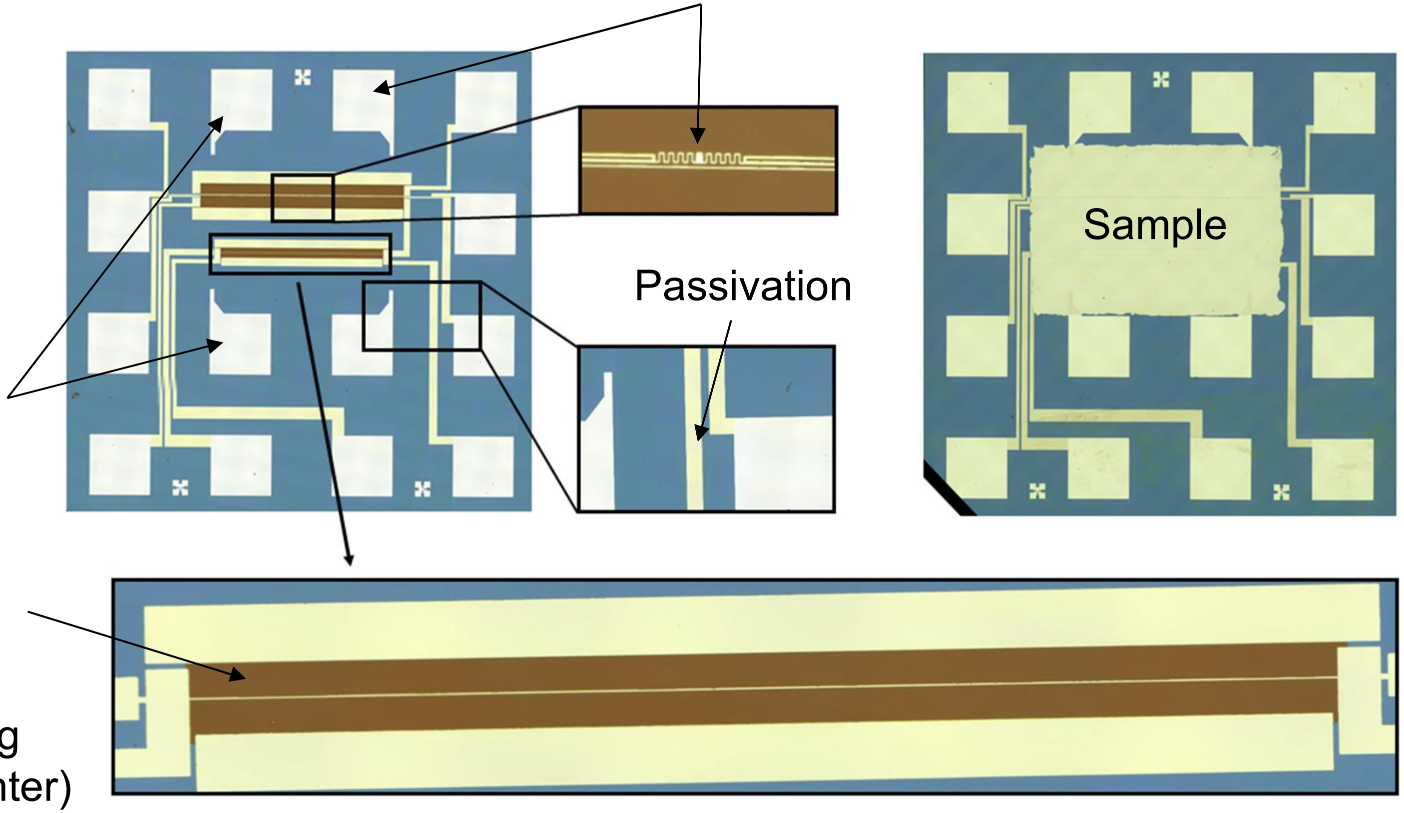
Sensor layout

Back side of the chip.

1. Seebeck measurement (thermometer with "hot contact" on membrane. Cold contact is "needle contact")

2. El. Conductivity & Hall measurement using 4-Point Van-der-Pauw (needle contacts)

3. Thermal conductivity (two suspended membranes with heating stripe aligned to the center)



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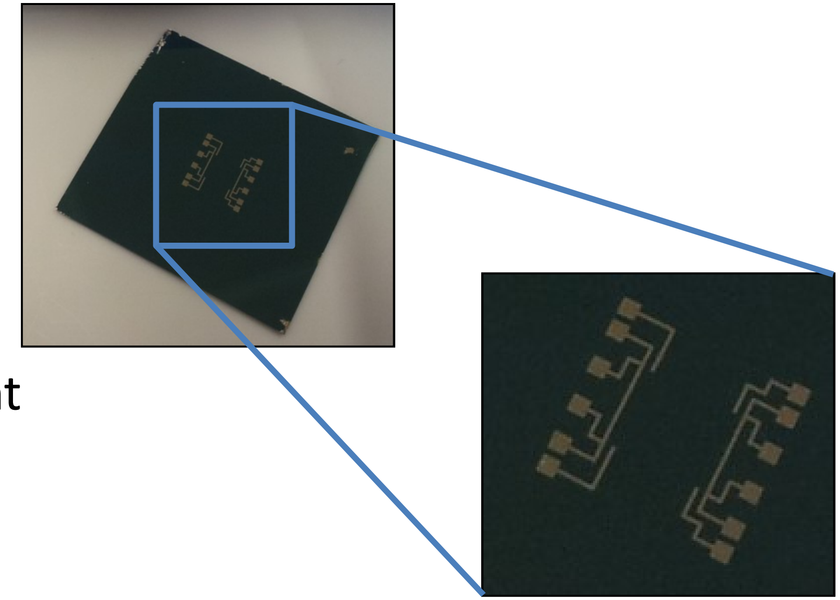

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Electrical conductivity measurement

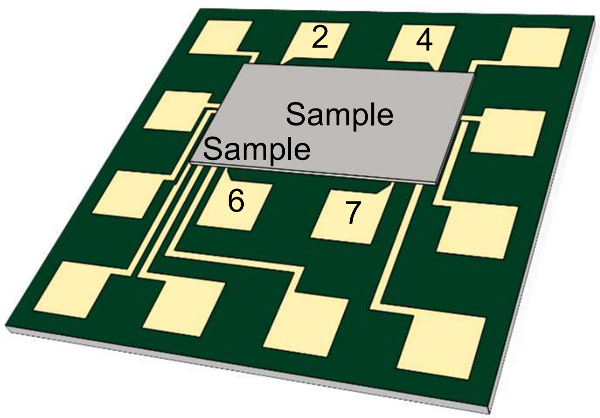
Using the Van-der-Pauw Method

- Very well known measurement technique
- Needs homogeneous layer thickness *t* (must be measured otherwise)
- Contacts must be small compared to sample
- Contact must be at the edge

→ can be fulfilled using the deposition mask

- Possible resistivity range up to $10^8\ \Omega$

$$ZT = \frac{S^2\sigma}{\lambda}T$$



sensor layout for VdP measurement

$$\exp\left(-\frac{\pi t}{\rho}\cdot R_{26,47}\right) + \exp\left(-\frac{\pi t}{\rho}\cdot R_{24,67}\right) = 1$$

Van-der-Pauw formula

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Hall constant measurement

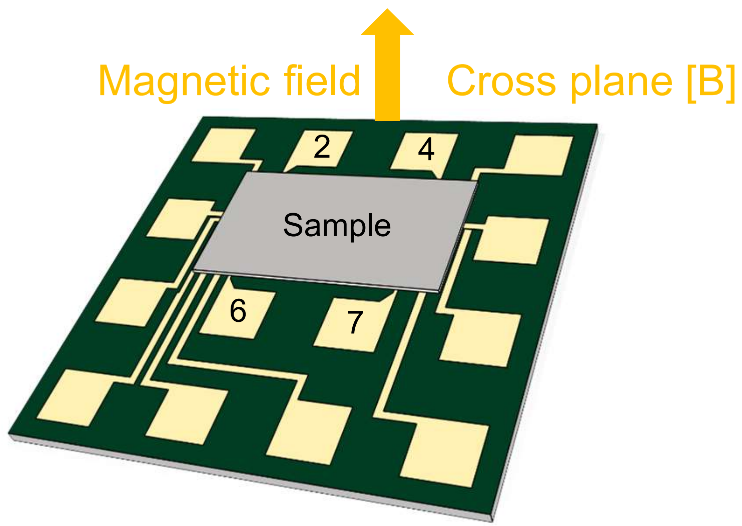
Using the Van-der-Pauw Method

- Very well known measurement technique
- Needs homogeneous layer thickness *t* (must be measured otherwise)
- Contacts must be small compared to sample
- Contact must be at the edge

➡ can be fulfilled using the deposition mask

- Possible resistivity range up to $10^8\ \Omega$

$$A_H = \frac{t}{B} \Delta R_{27,46}$$



$$\Delta R_{27,46} = R_{27,46(B=1)} - R_{27,46(B=0)}$$

➡ Hall mobility, carrier concentration

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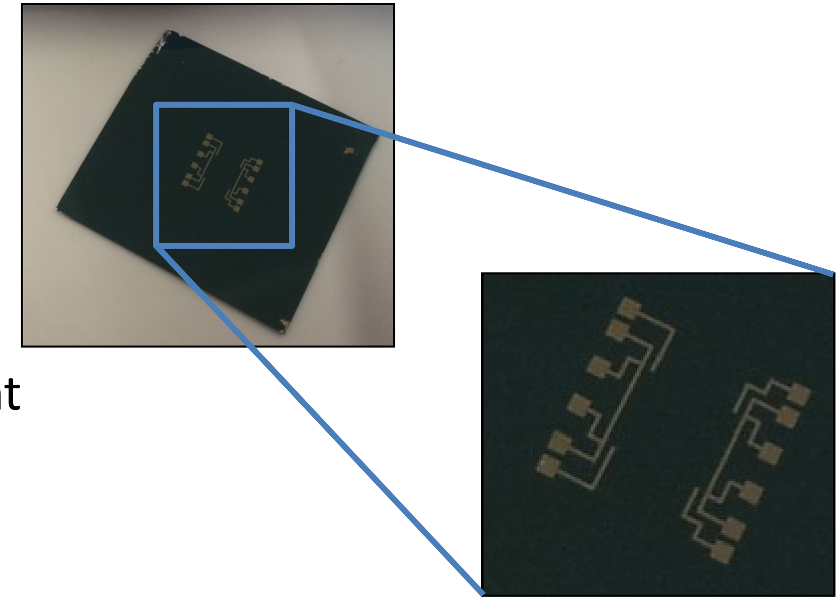
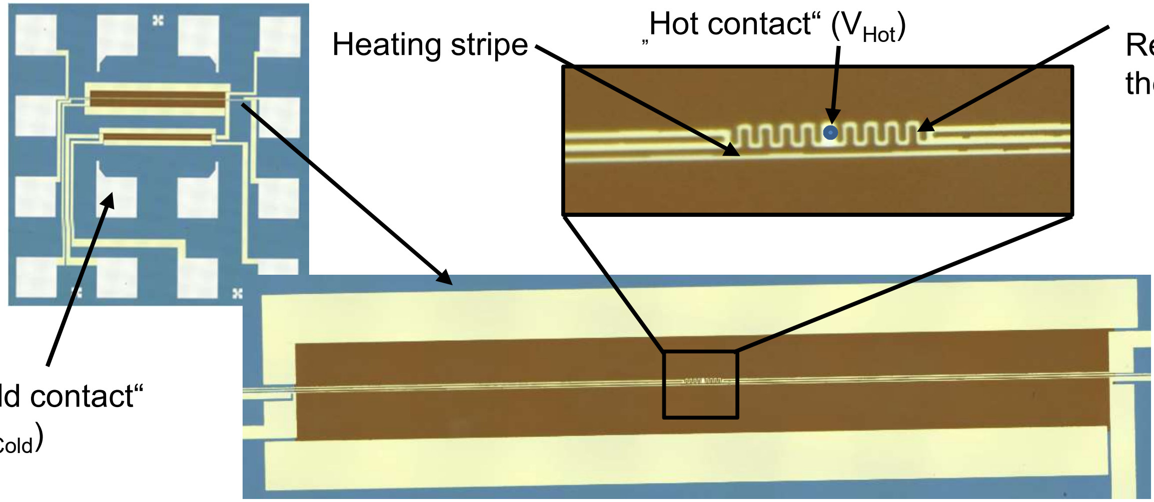
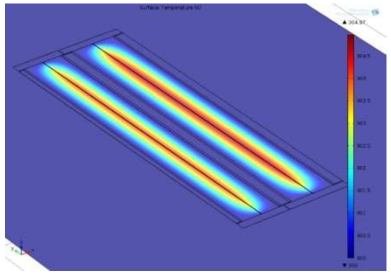


Seebeck coefficient measurement

- Heater and thermometer on membrane
  - ➡ Large gradients are possible
  - ➡ Fast measurement (short thermal relaxation time)
  - ➡ Temperature gradient can be adjusted with I_{heater}

$$ZT = \frac{S^2\sigma}{\lambda}T$$





Membrane temperature distribution with heating stripe located in the middle

$$With\ T_{Hot} \propto \frac{V_{RT}}{I_{RT}}\ \&\ T_{Cold} = T_0\ and\ V_{Th} = V_{Hot}\text{-}V_{Cold}$$

➡

$$S = \frac{-V_{\mathrm{Th}}}{\Delta T}$$

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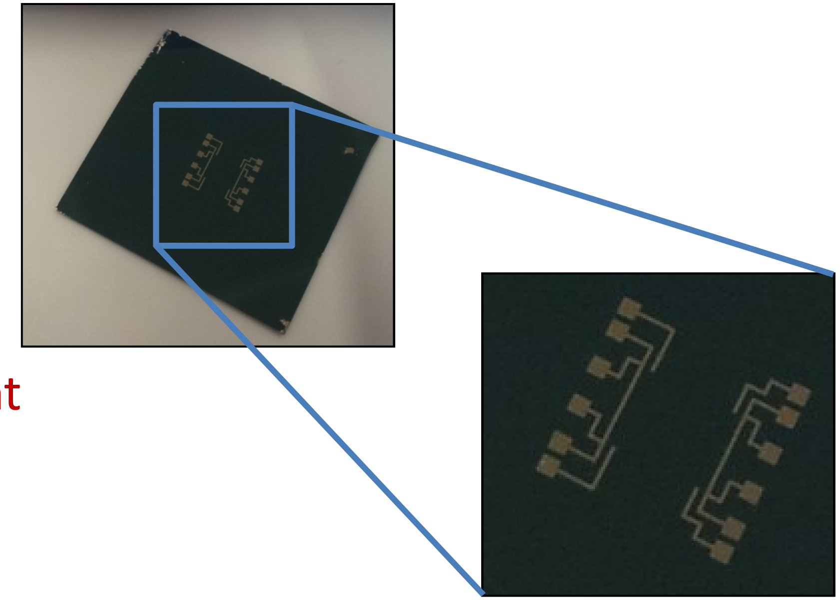


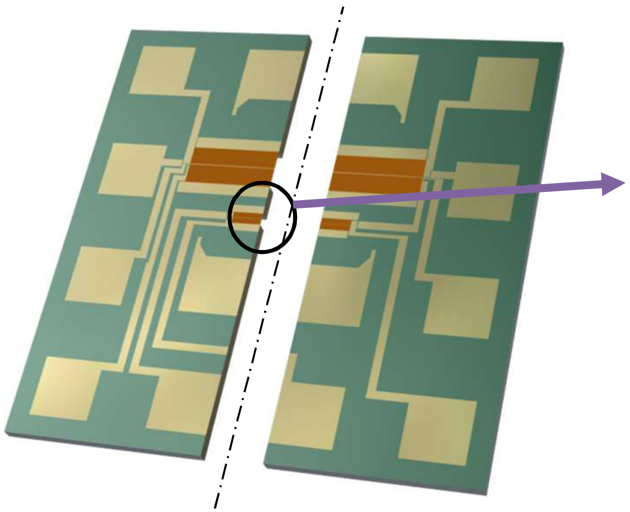
Thermal conductivity measurement

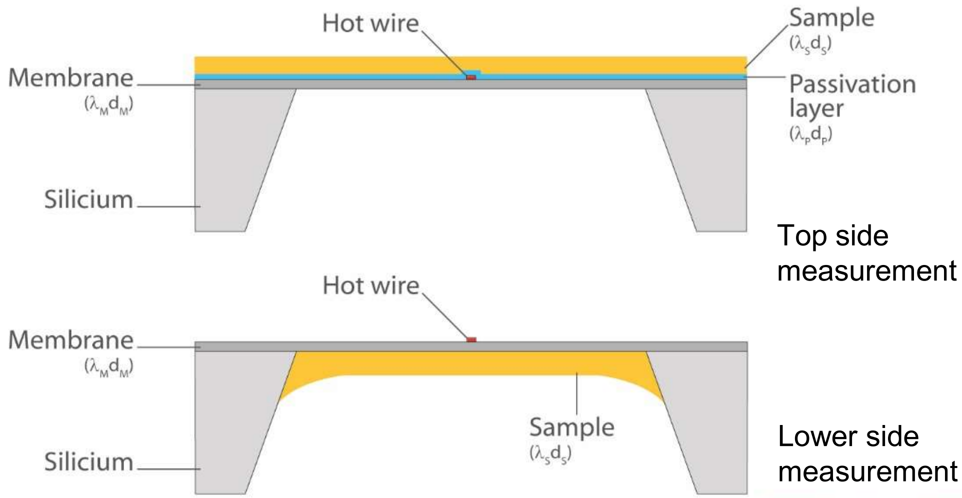
- Determination of the thermal conductivity using a hot stripe method
  ➡ Hot stripe acts as heater and sensor at the same time

- Two membrane setup for heat loss correction due to radiation.

$$ZT = \frac{S^2\sigma}{\lambda}T$$

- 3ω measurement (transient measurement, additional specific heat measurement is possible)





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Thermal conductivity measurement

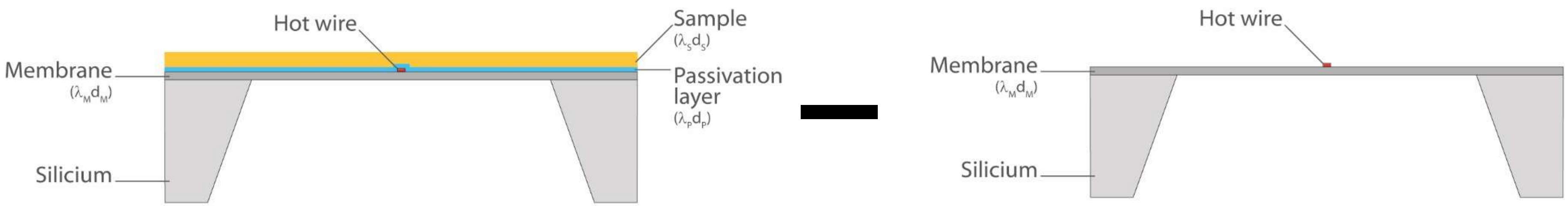
- Determination of the thermal conductivity using a hot stripe method

  Hot stripe acts as heater and sensor at the same time
- Two membrane setup for heat loss correction due to radiation.
- 3ω measurement (Transient measurement, additional specific heat possible)
- Differential measurement (zero curve correction)

$$ZT = \frac{S^2\sigma}{\lambda}T$$

Sample measurement

Empty sensor measurement



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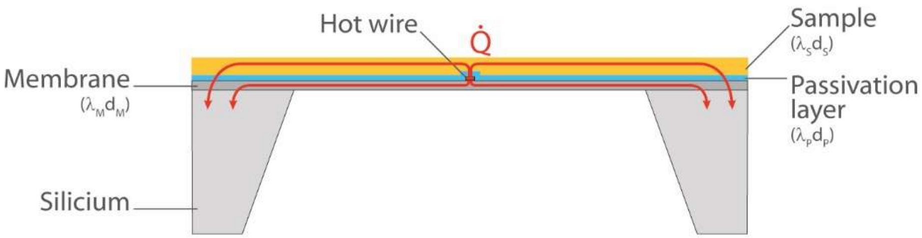


1. *Measurement of R_{hot_wire} (unheated)*
2. *Joule Heating (heating Power P = I²*R)*
3. *Heat flows through membrane & sample into Si-Rim*
4. *Measurement of R_{hot_wire} (heated)*

➡ $Total\ th.\ conductance = \frac{heating\ Power}{\Delta T}$

Thermal conductivity measurement

- Heating with known heating power P and measure mean temperature increase ΔT_M

➡ Thermal conductivity can be calculated using the geometry data of the membrane

- To correct the influence of heat loss due radiation, a two membrane setup + shielding is used

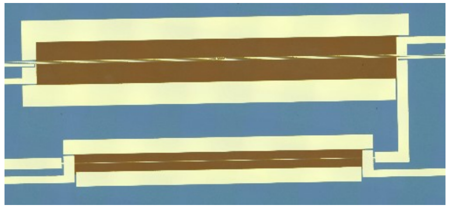
➡ Additional information about emissivity

Solved 1-D heat flux equation

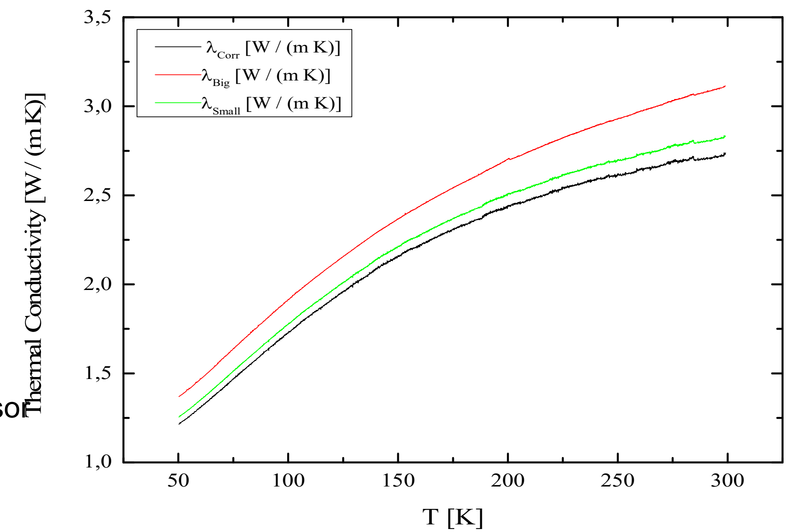
$$G\left[1-\frac{2}{\upsilon l}\tanh(\upsilon\frac{l}{2})\right]=G_m(\lambda,\varepsilon)+G_h(\varepsilon_h)$$

with $$G=\frac{P}{\Delta T_M}$$

and $$\Delta T_M=\frac{R(T_0+\Delta T_M)-R(T_0)}{\beta\cdot R(T_0)}$$



Two membrane setup on the sensor



Thermal Conductivity measurement of 100 nm Si_3N_4

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Völklein, F., Reith, H., and Meier, A. 2013. Measuring methods for the investigation of in-plane and cross-plane thermal conductivity of thin films. *physica status solidi (a)* 210, 1, 106–118.

Thermal conductivity measurement

Q: “What's the “minimum” thickness to measure?“

A: Major point is: „$\lambda_S d_S \geq 2 \cdot 10^{-7}\ \mathrm{WK}^{-1}$ “

The thermal conductance of the sample should be in the same range or bigger than the thermal conductance of the membrane (empty chip), as we use a differential measurement.

Material (thin film)	Th. Cond. @ RT	Min. thickness	Max. thickness
Metal	~ 100 W/mK	5 nm	0,25µm
TE material	~ 3 W/mK	50 nm	1 µm
Organic material	~ 0.4 W/mK	400 nm	15 µm

Measurement range of the thermal conductivity setup for different material classes at room temperature. Journal of ELECTRONIC MATERIALS; https://doi.org/10.1007/s11664-017-5989-4 2017 The Minerals, Metals & Materials Society

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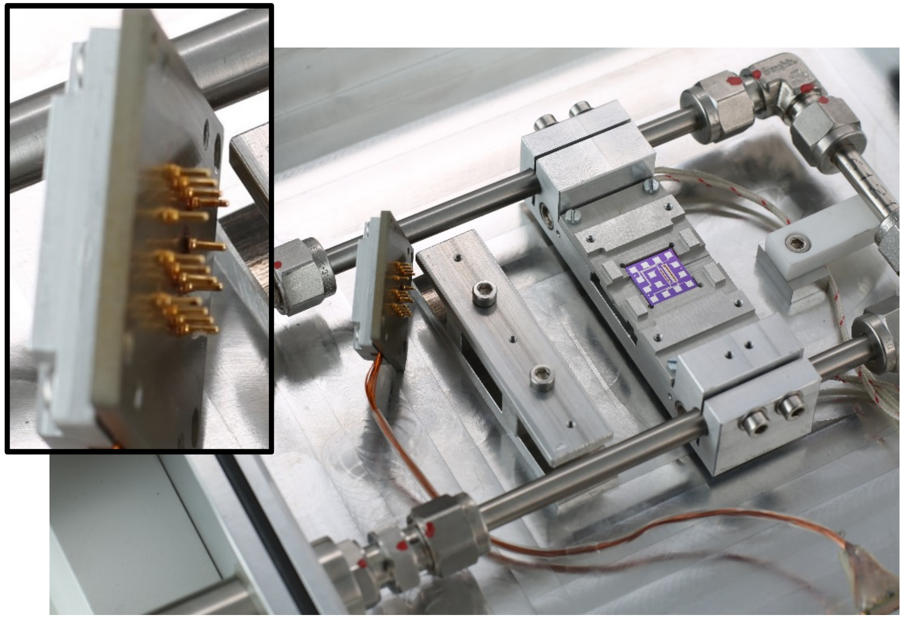
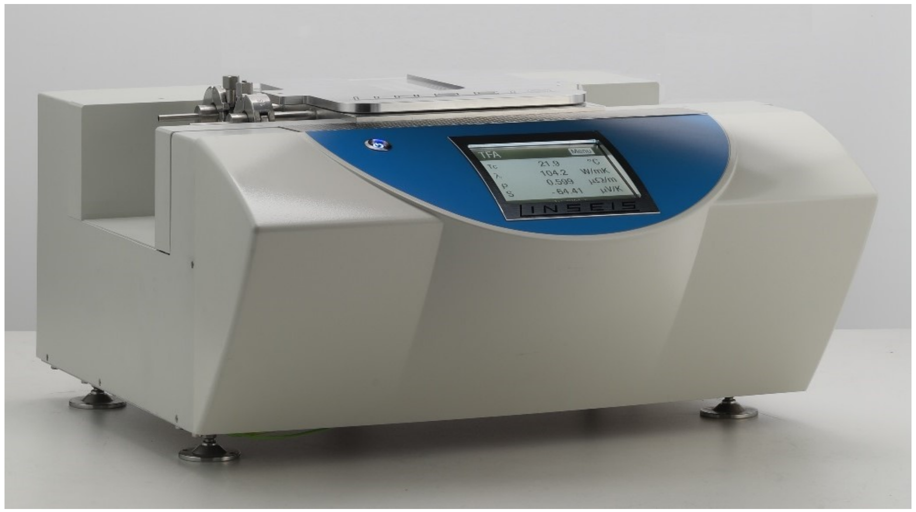
Measurement setup (without Hall-option)

- Modular design
- Easy chip installation and connection (spring pins, no bonding)
- Optimized thermal and magnetic behavior (can be controlled fast and easily)
- Chamber is designed for vacuum applications (10^{-5} mbar)





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Measurement Setup (with Hall option)

- Complete measurement device consists of
  - Measurement chamber
  - Rack, including electronics
    - Thyristor
    - Lock-in amplifier
    - Electronics
  - Vacuum pump
  - Moveable electromagnet including chiller
  - KREG (LN_2 Cooling)
    - Dewar
    - Valve & control box

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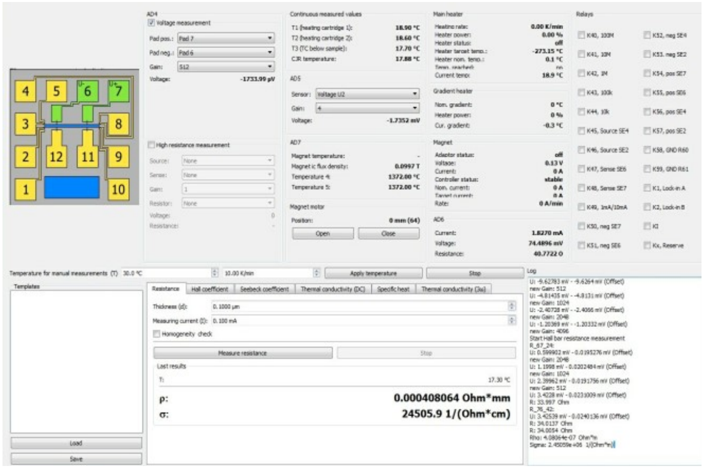
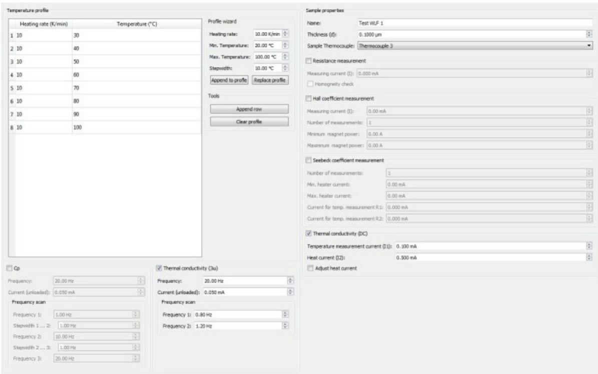
Measurement Software

- Temperature and magnetic field regulation (swipes)
- Optimized electronics for the measurement tasks
  - Basic device
  - 3 ω unit
  - Magnet control unit
- Current values (sensors)
- Measured values (samples raw data)
- Automatic measurements
  - Heat profiles
  - Measurement tasks
- Regulation of measurement properties.



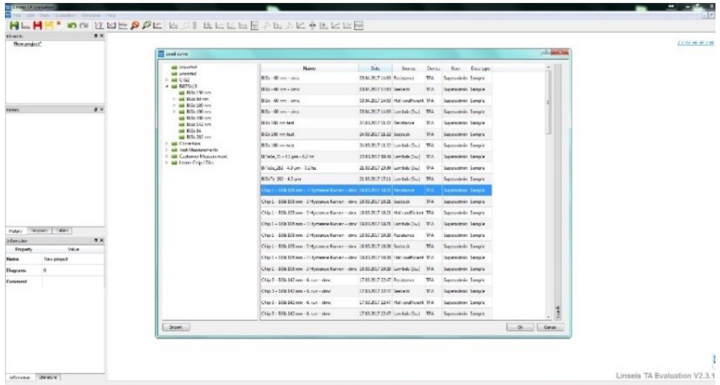
Manual measurement & current values



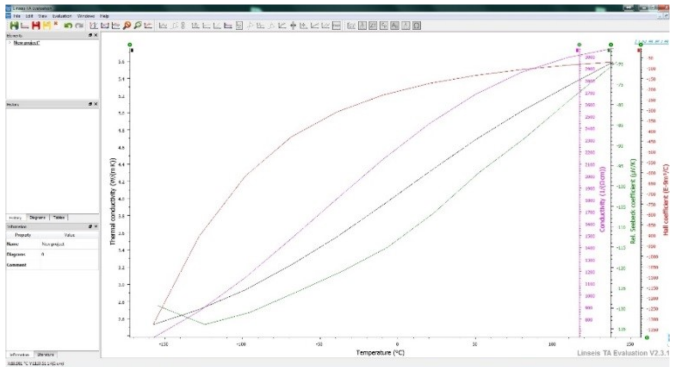
Automated measurements & profiler.

Evaluation Software

- Database for raw data management
- Evaluation of the data with integrated plugins
  - Thermal conductivity
    - Zero curve correction
    - Specific heat calculation
    - Emissivity (dual membrane measurement)
  - Seebeck coefficient
    - Single point evaluation
    - Slope method evaluation
    - Average evaluation per temperature point
  - Resistivity / el. conductivity
  - Hall coefficient
  - Hysteresis (separation of heating / cooling)
  - ZT calculation / Powerfactor calculation
- Plotting (time / temperature)
- Export data



Database for raw data management



Raw data evaluation and data post-processing.

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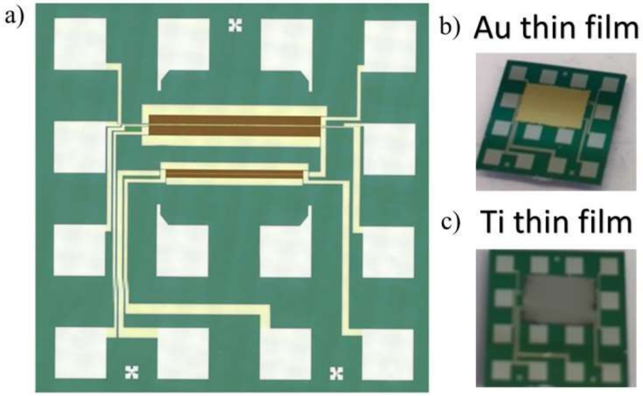
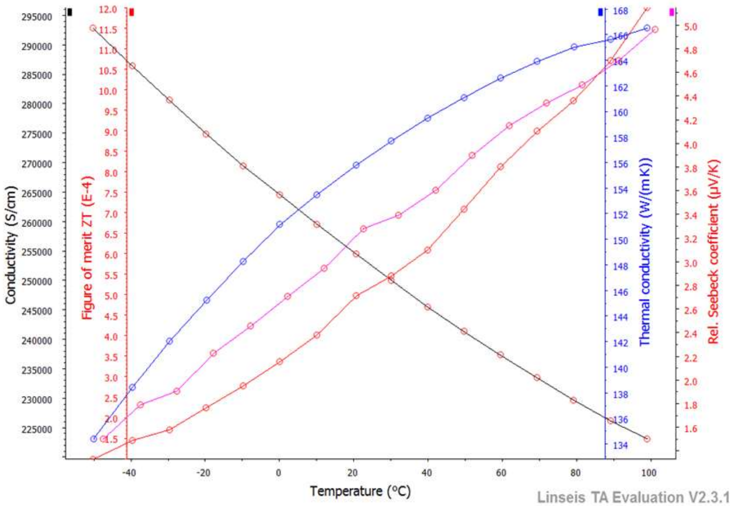
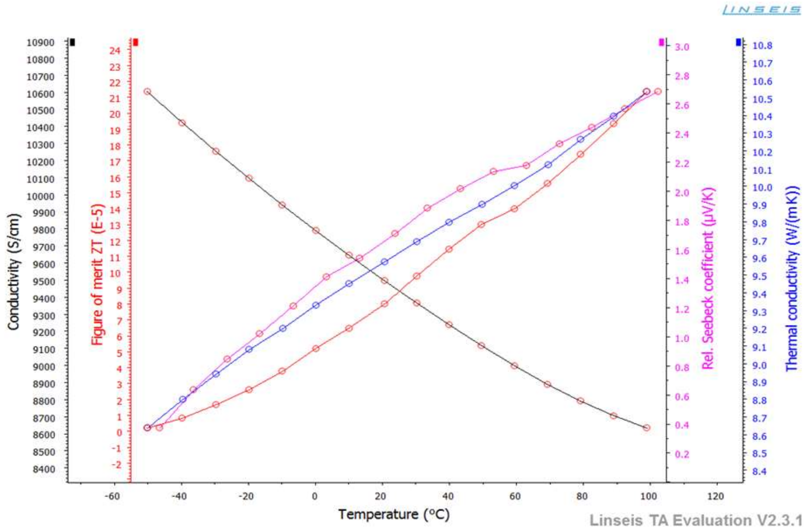


Au 100 nm sputtered:

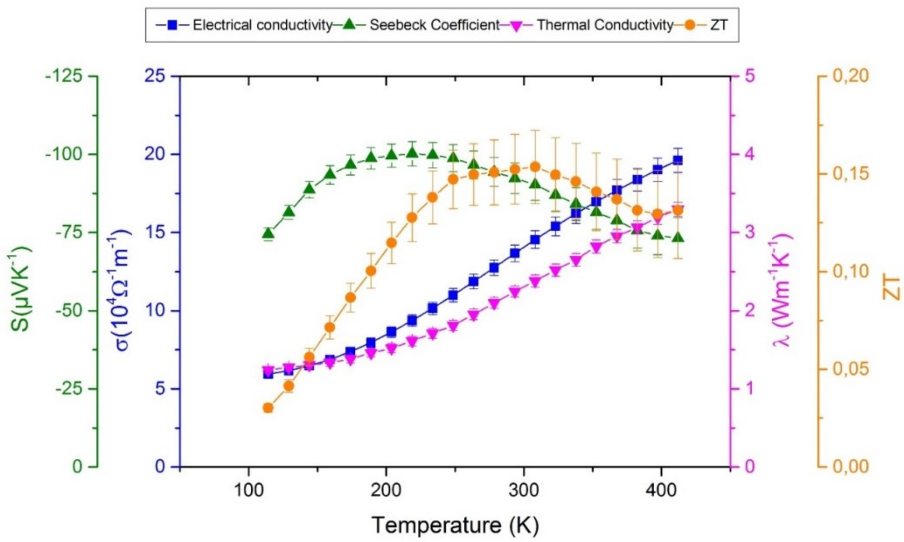


Ti 100 nm sputtered:

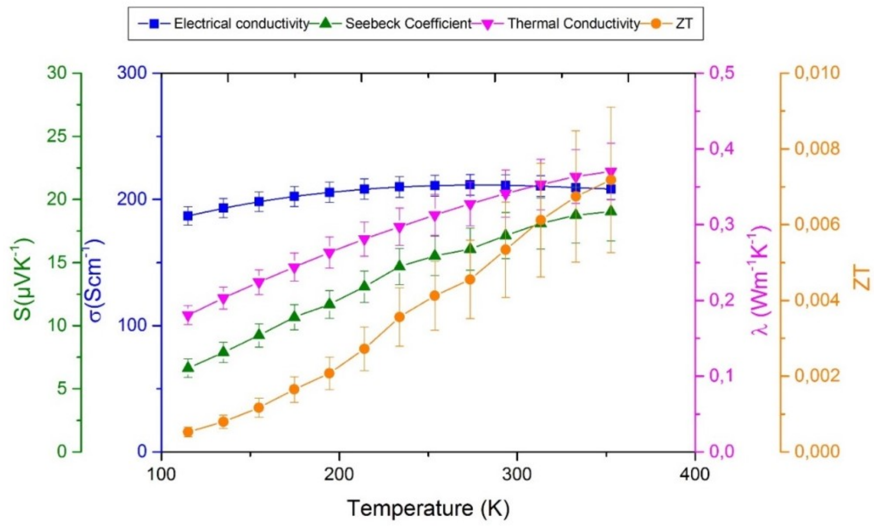


Applications

84 nm $Bi_{87}Sb_{13}$; evaporated



15 µm PEDOT:PSS; drop casting



Outline

1. Motivation & Project Goals
2. Concept
3. Chip layout
    1. El. conductivity measurement
    2. Hall constant measurement
    3. Seebeck coefficient measurement
    4. Thermal conductivity measurement
4. Measurement setup
5. Applications
6. Summary

Short summary

Unique commercially available measurement instrument for thermo-physical properties of thin films:

- -170 up to 300°C
- Magnetic field up to ± 1 Tesla
- Vacuum up to 10^{-5} mbar
- Ready-to-use disposable sensor for the following parameters (in-plane):
  - ρ/σ electrical resistivity/conductivity
  - S Seebeck coefficient
  - λ thermal conductivity (3ω method)
  - Cp specific heat capacity
  - ε emissivity
  - A_H Hall constant (n + μ charge carrier concentration and mobility)

➡ TFA - Thin Film Analyzer
available from Linseis Messgeräte GmbH

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Thank you for your attention!

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Any questions?