DOE/NSF Thermoelectric Partnership
Project SEEBECK
Saving Energy Effectively By Engaging in Collaborative research and sharing Knowledge

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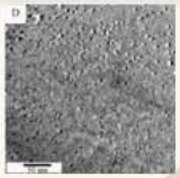
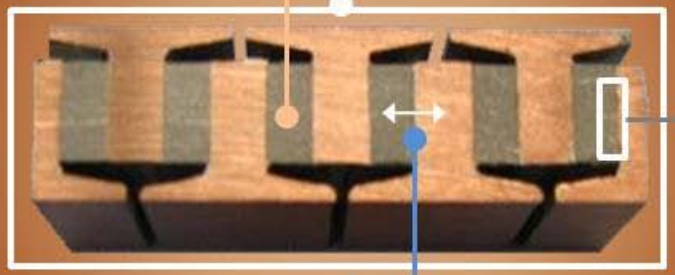
Ack. Audrey Chamoire, Christopher M. Jaworski
Presentation:
2011 Thermoelectrics Application Workshop, San Diego, Jan 3-6, 2011

Structure of this project

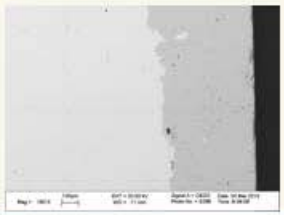
1. Materials
NU – OSU
PbSe –based or
Mg_2X (X=Ge,Si,Sn,Pb)



2. Thermal Management
BSST
Innovative
Design and Architecture



3. Interfaces
VT – ZTPlus
New flexible
Ag Nanopaste and metallization



4. Metrology
BSST – OSU – NU
VT – ZTPlus
(ITE)
Chemical and Physical characterization

5. Durability
BSST – OSU – NU
VT – ZTPlus
Mechanical and Thermal shock resistance

1. Materials

1.1 PbSe

D. Parker, D. J. Singh " High-temperature thermoelectric performance of PbSe", 29th International Conference on Thermoelectrics, Shanghai, China, May 30-June 3, 2010

- 1.1.1 Resonant levels
- 1.1.2 Nanostructuring

1.2 $Mg_2(Si, Sn)$

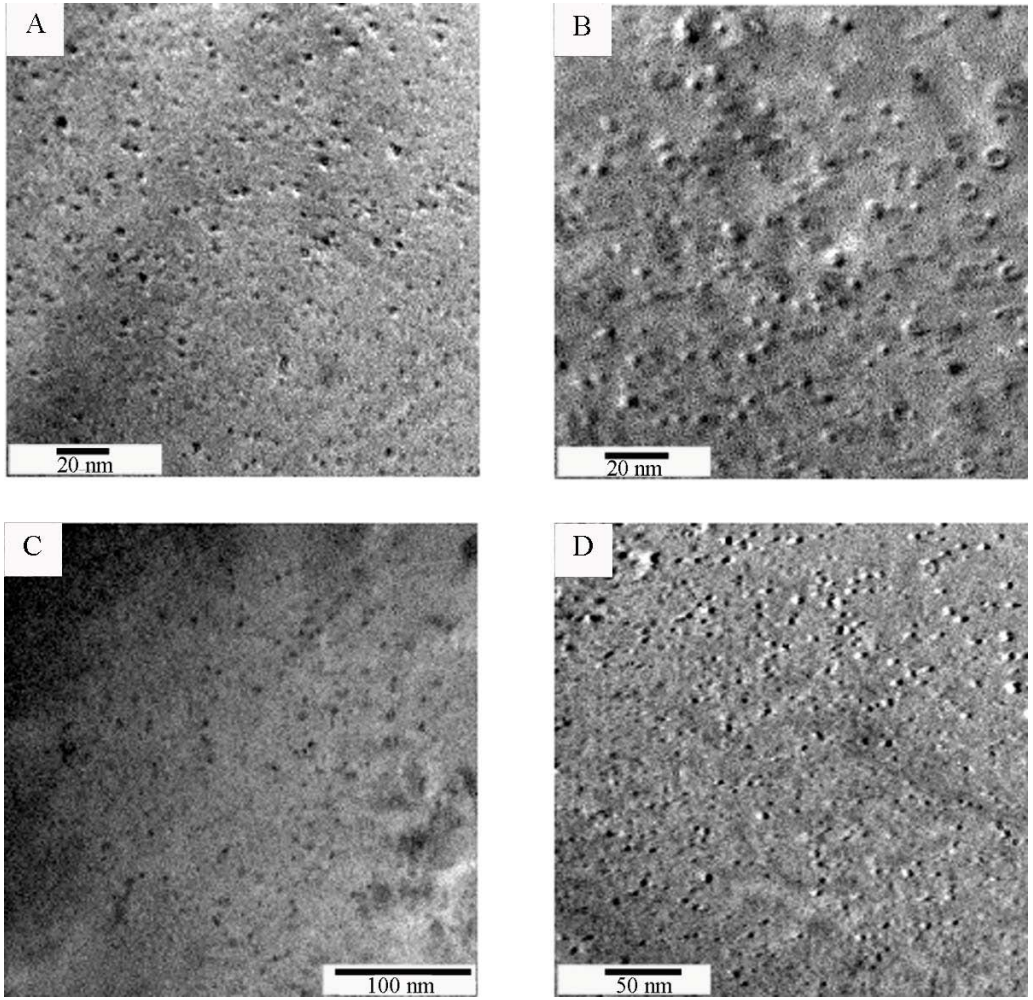
- 1.2.1 Resonant levels
- 1.2.2 Nanostructuring

Northwestern, OSU

1.1.2 Nanostructuring PbSe

Same bulk solid-state techniques as PbTe

J. R. Sootsman, R. J. Pcionek, H. Kong, C. Uher, M. G. Kanatzidis, "Strong Reduction of Thermal Conductivity in Nanostructured PbTe Prepared by Matrix Encapsulation", Chem. Mater. ***18*** *p.4993 (2006).*



Dispersed nanoparticles in samples of Sb-doped PbTe:
(A)PbTe-Sb(2%)
(B)PbTe-Sb(4%)
(C) PbTe-Sb(8%) and
(D) PbTe-Sb(16%).

Initial results on PbSe:
ZT~0.8

1.2.1 Resonant levels in $Mg_2(Si,Sn)$

1. Need details of band structure to choose the right level

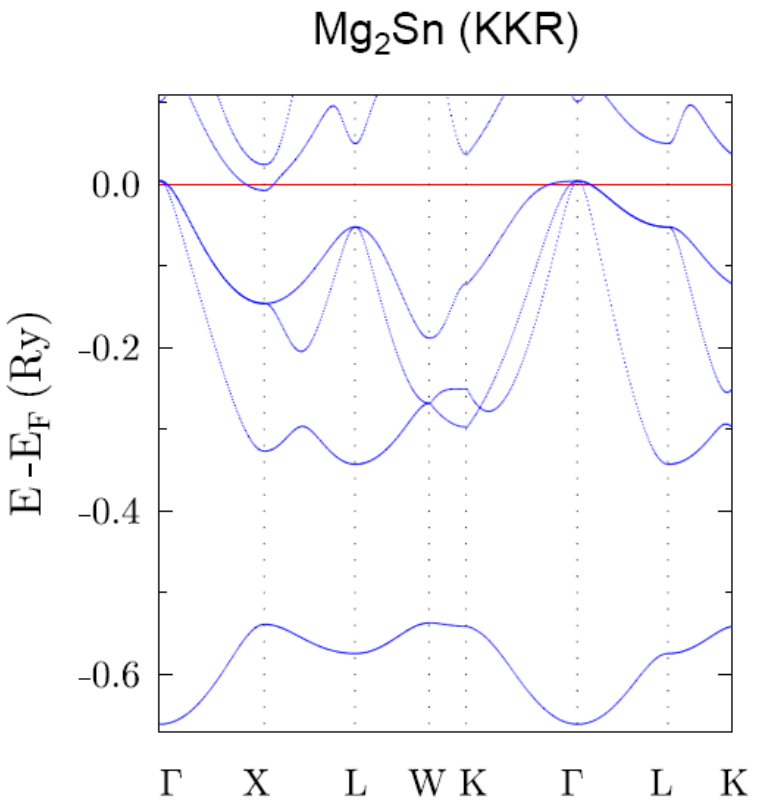
2. Start from band structure calculations

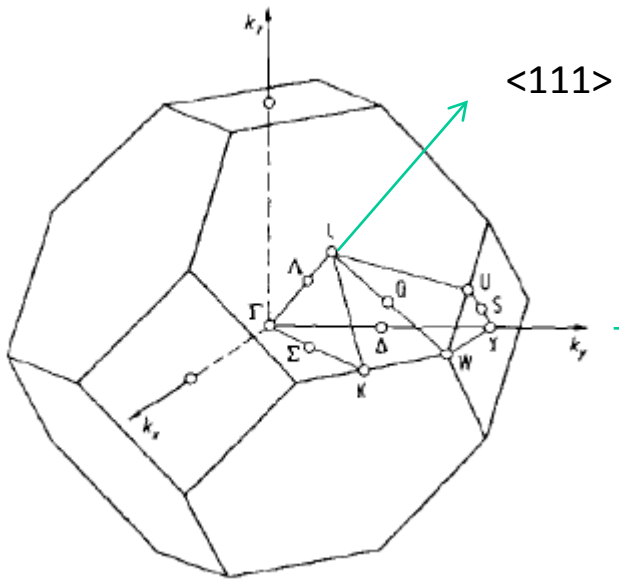
   example(J. Tobola): Ag creates additional DOS in valence band

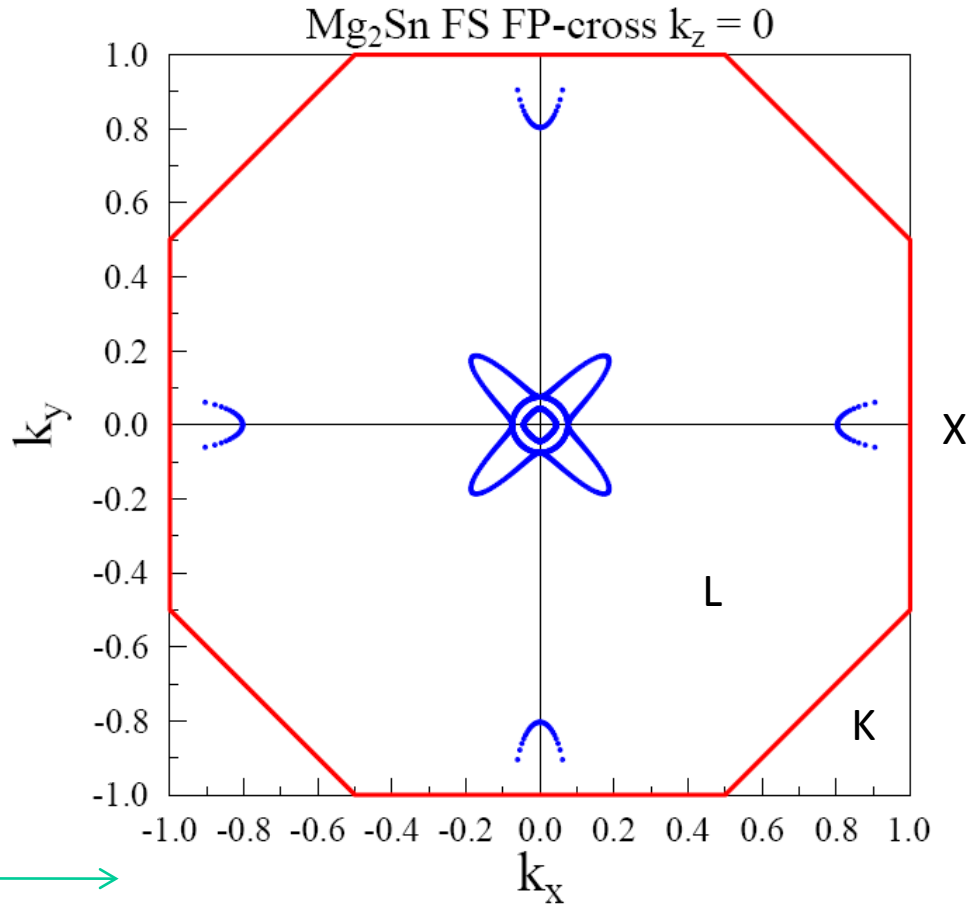
3. Preliminary experiments: Shubnikov-de Haas measurements on single-crystal Mg_2 Sn doped with Ag

Christopher M. Jaworski[1] , H.Y. Chen[2], N. Savvides[2], B. Wiendlocha[3], J. Tobola[3], J.P. Heremans[1,4] Presented at the 2009 International Conference on Thermoelectricity

Calculated Fermi Surface

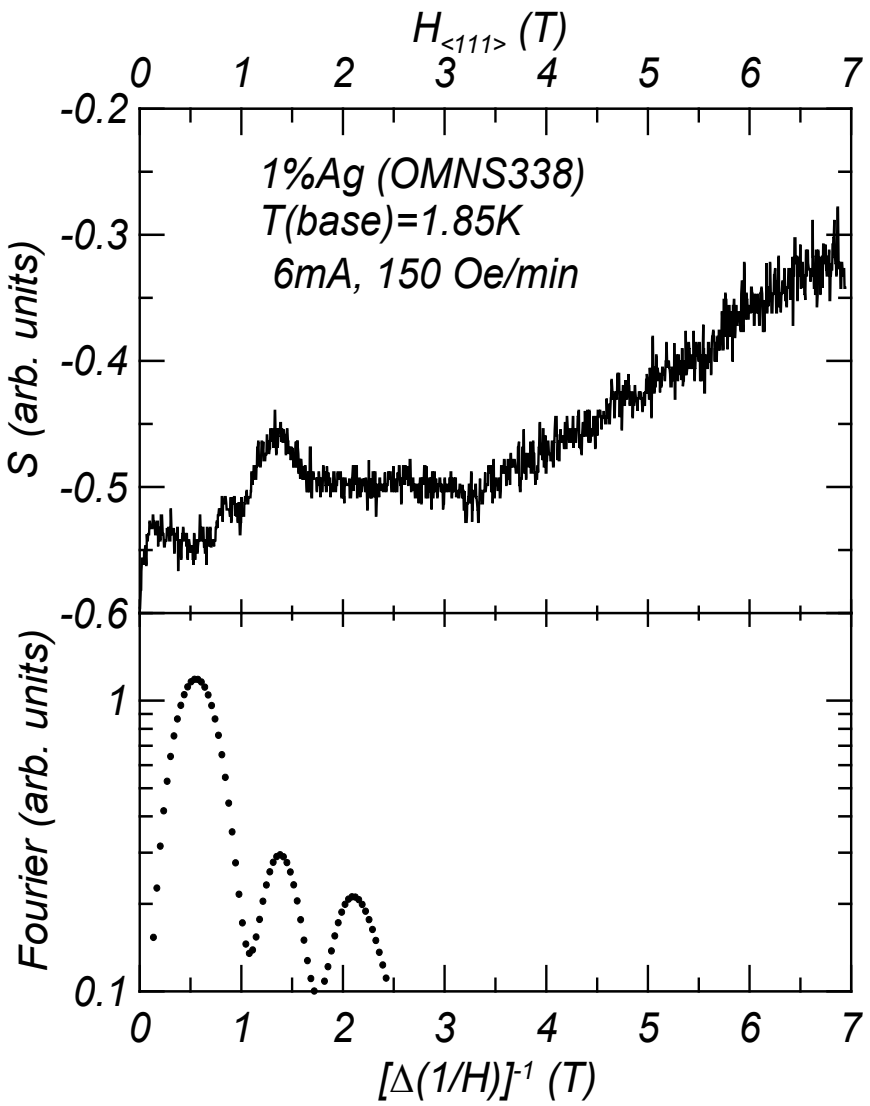


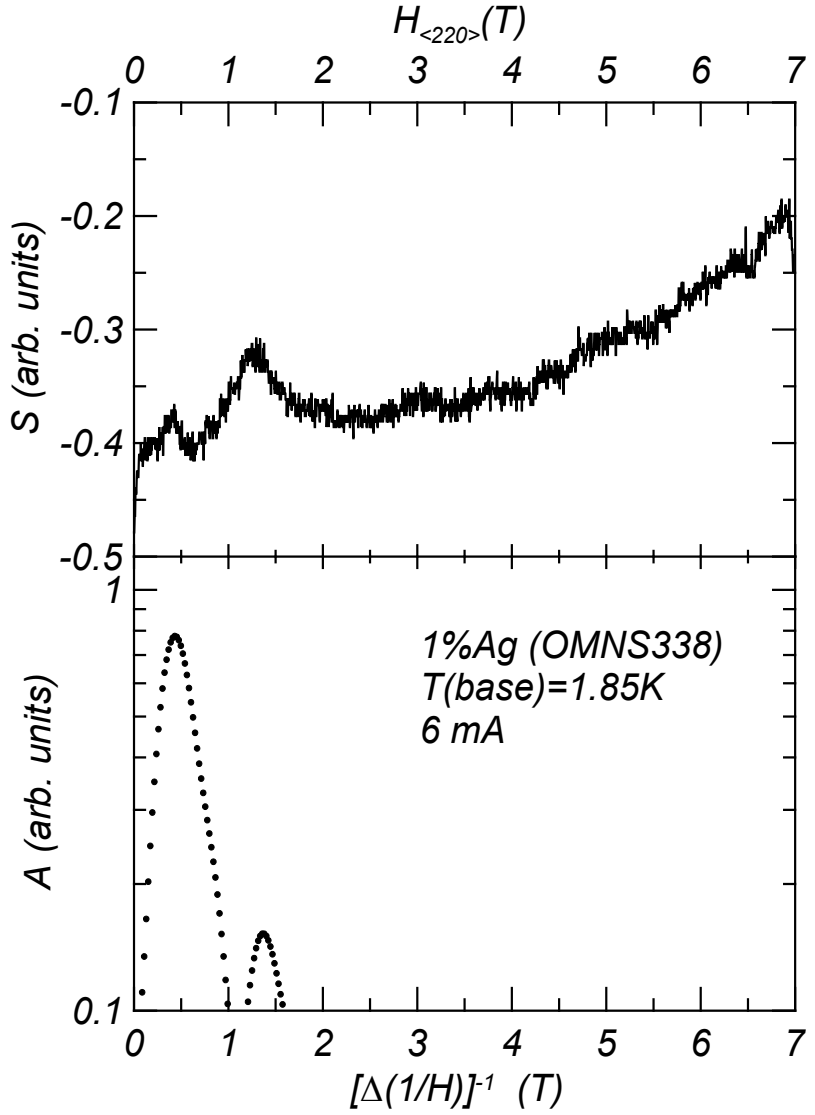




J. Tobola, Krakau, Poland

Representative Traces 1 % Ag





SdH Oscillations

Ag content (at %)	0.25%	1%
$1/\Delta(1/H)$ (Tesla), H//<111>	1.24	0.61
$A_{F,<111>}$ (m^{-2})	1.18×10^{16}	0.58×10^{16}
$1/\Delta(1/H)$ (Tesla), H//<111>		1.3
$A_{F,<111>}$ (m^{-2})		1.2×10^{16}
$1/\Delta(1/H)$ (Tesla), H//<220>	0.65	0.61
$A_{F,<220>}$ (m^{-2})	0.62×10^{16}	0.58×10^{16}
$1/\Delta(1/H)$ (Tesla), H//<220>	1.25	1.378
$A_{F,<220>}$ (m^{-2})	1.19×10^{16}	1.31×10^{16}

Possible cyclotron masses from SdH and $E_F(S)$

$$m_c = \frac{\hbar^2 A_F}{\pi 2 E_F}$$

m_c (.25% Ag) = 0.013, 0.025 m_e
m_c (1% Ag) = 0.005, 0.011 m_e
Same result for <111> <220>

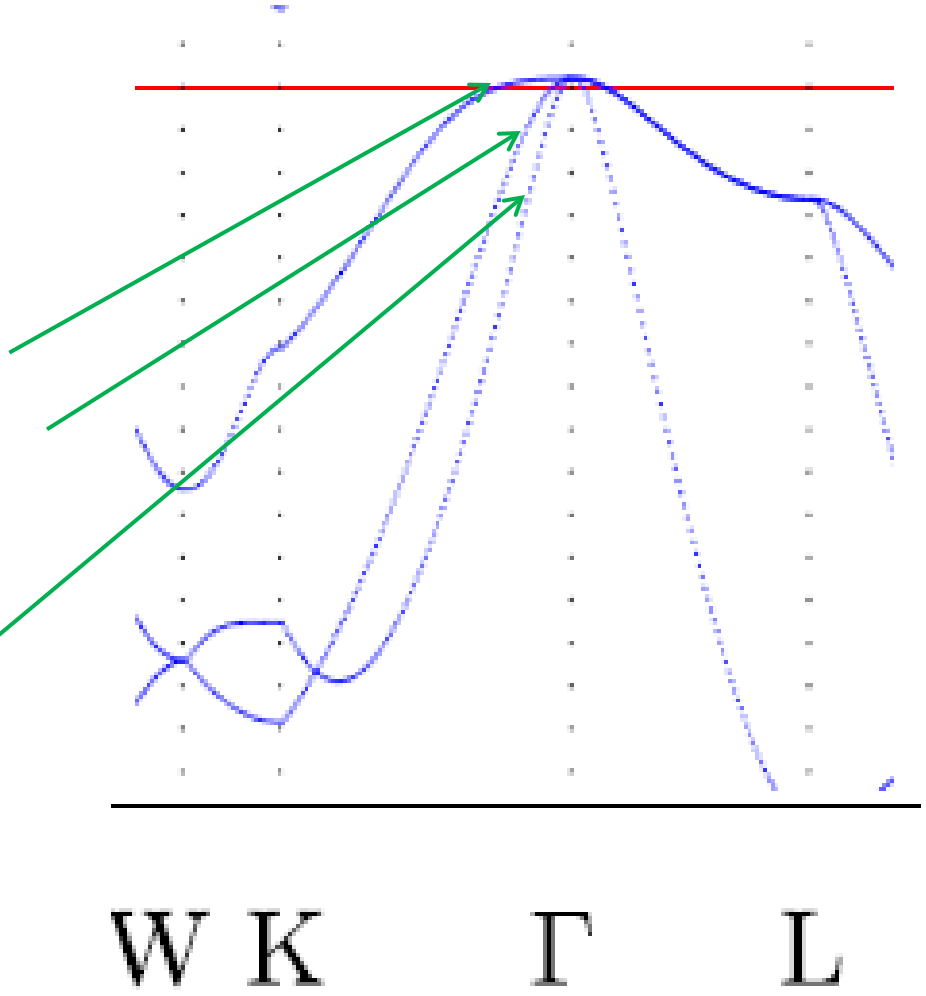
SdH and calculated masses

Γ-K direction - three bands crossing the Fermi level,

0.9-1.2 for external band,

0.11-0.15 for intermediate one

0.03-0.05 for the internal band



1.2.2 Nanostructuring $Mg_2(Si,Sn)$

Same technique: liquid encapsulation

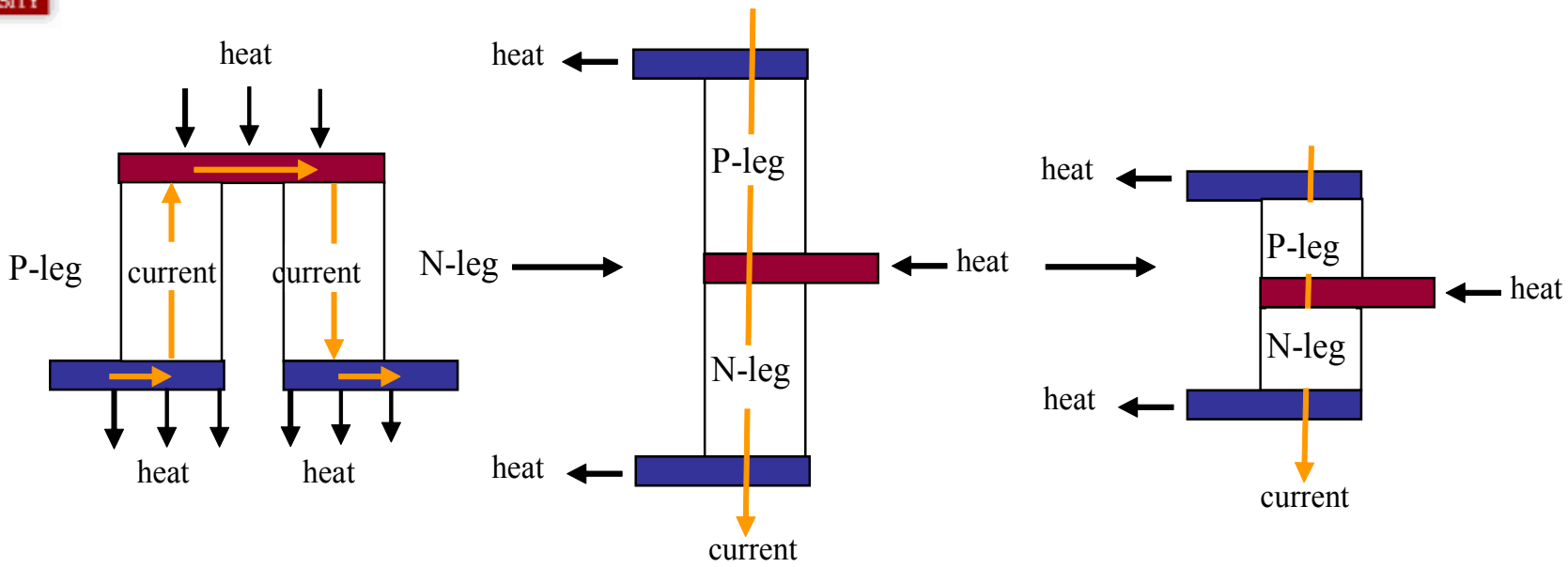
$Mg_2 Si_{1-x}Sn_x$ /Y, $Mg_2Si_{1-x}Pb_x$/Y and $Mg_2Sn_{1-x}Pb_x$/Y systems

Y as element

Y as compound that do not form compounds with Mg_2X :

- binary silicides
- Binary stannides

2. Thermal Management: new BSST designs



Heat / electrical current in cross-flow geometry

(1)reduced number of interfaces and components
=>reduced parasitic losses

(2)high power density materials => much smaller sizes

(3)segmentation across the specified temperature differential to maximize efficiency

Will be adapted to high-ZT materials & reduced thickness

BSST

3. Interfaces

3.1 TE material metallization

3.1.1 Co-pressing of metallized layers and powders during process of Spark Plasma Sintering (SPS)

3.1.2 Post processing of annealed samples and deposition of metals using Physical Vapor Deposition (PVD) methods.

3.2 New compliant device interconnects

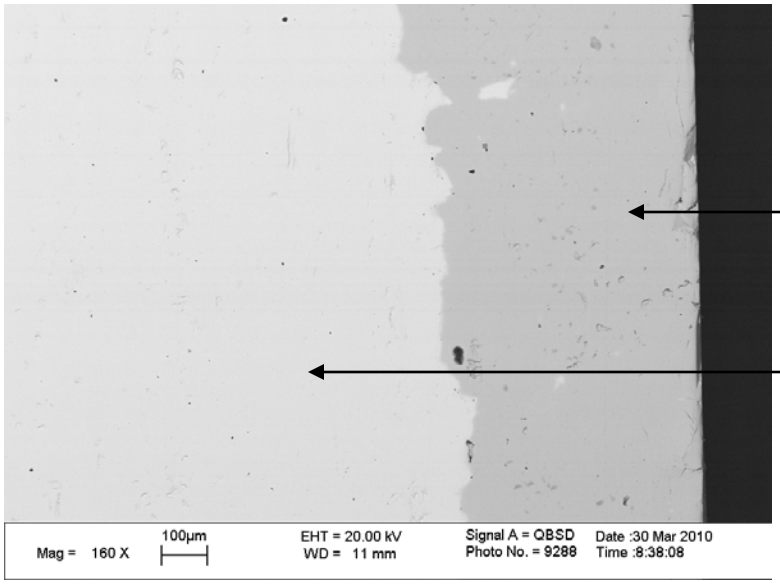
3.2.1 Nanosilver

3.2.2 New systems

Virginia Tech, ZT Plus

3.1 TE material metallization

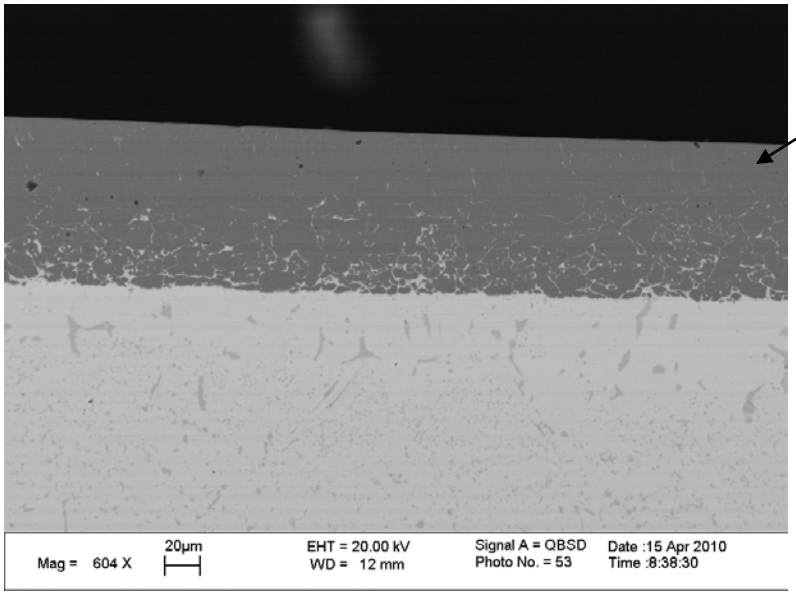
Direct co-sintering during material consolidation (SPS)



400µm layer of SnTe

p-type PbTe

from powders during Spark Plasma Sintering.



Fe foil co-sintered with p-type PbTe

interdiffusion and likely a eutectic structure

Issues: interdiffusion, diffusion barriers

3.2 Nanosilver interconnect packaging material

Pb-free, high-T die-attach solution:

- Joint formed below 280°C;
- Melting at 960°C;
- Thermal conduct > 150 W/m-K.

Binder
Surfactant
Thinner

Surfactant + Organic thinner + Ag nano-powder

Silver Paste
nanoTach®

Uniform Dispersion

Heat up

Sintered joint

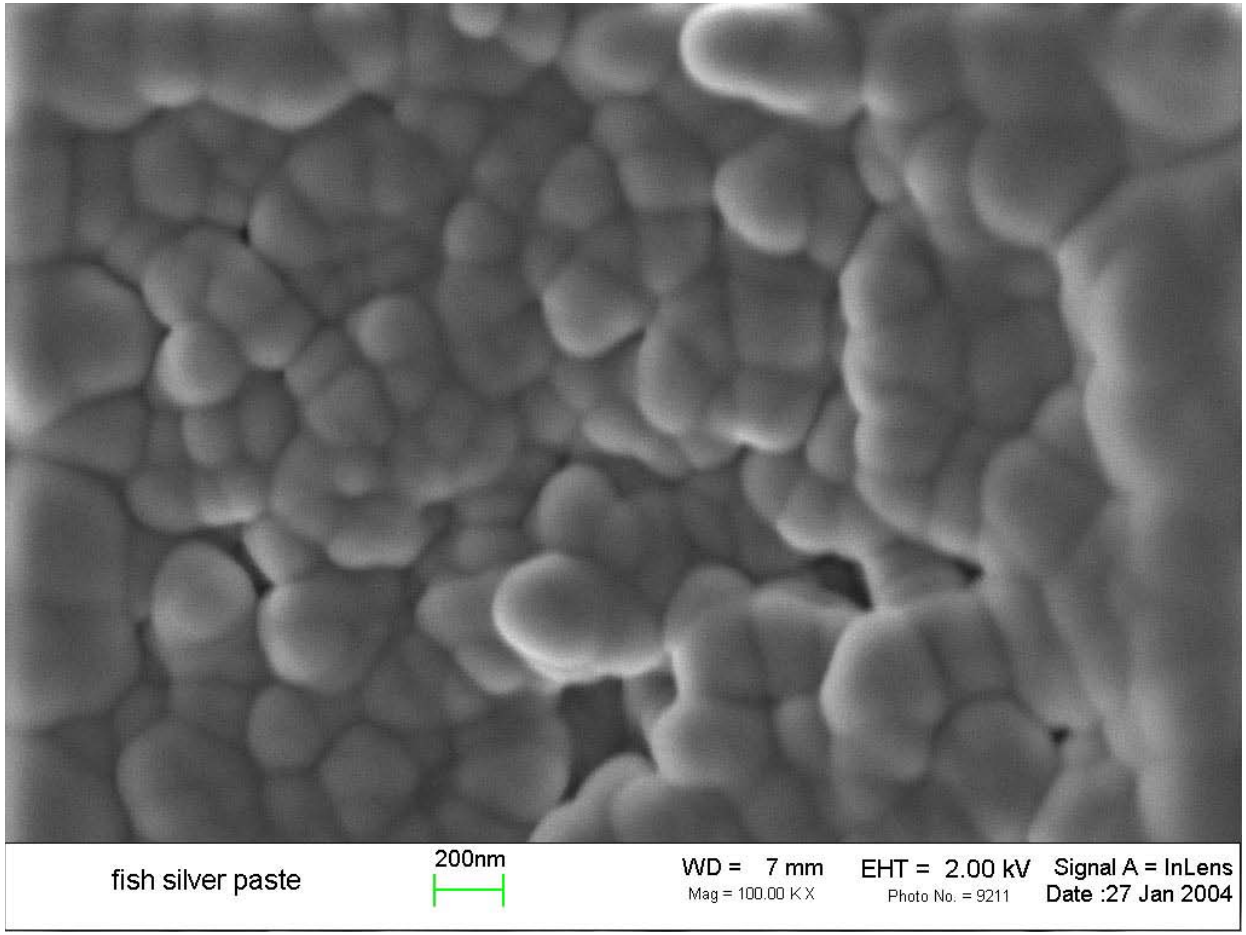
Applications:

Substrate

Cool down

Densification by diffusion

in situ nanosilver sintering in a scanning electron microscope



30 nm Ag Powder

15

Properties of nanosilver sintered die-attach compared with the others

	Processing temperature	Max. use temperature	Electrical conductivity 10^5 $(\Omega\text{-cm})^{-1}$	Thermal conductivity (W/K-cm)	Die-shear Strength (MPa)
Lead-tin solder	217°C	< 183°C	0.69	0.51	35
Lead-free solder	260°C	< 225°C	0.75	0.70	35
Gold-tin solder	310°C	< 280°C	0.625	0.58	30 - 60
Silver epoxy	100 – 200°C	< 200°C	0.1	0.1	10 – 40
High-Pb solder	340°C	< 280°C	0.45	0.23	15
Hysol® QMI 3555 R	300 – 450°C	<250°C	6.7×10^{-7}	0.80	20
		<280°C		<0.8	
Nano-Ag	< 275°C	< 961°C*	3.8	2.4	20 – 40

Nanosilver: developments needed

1. Adapt to thermoelectric materials

Difficulties: Ionic conductivity of silver
especially in chalcogenides

2. Less expensive alternatives

4. Metrology

4.1 TE material characterization exists
OSU, NU, ZTPlus, BSST

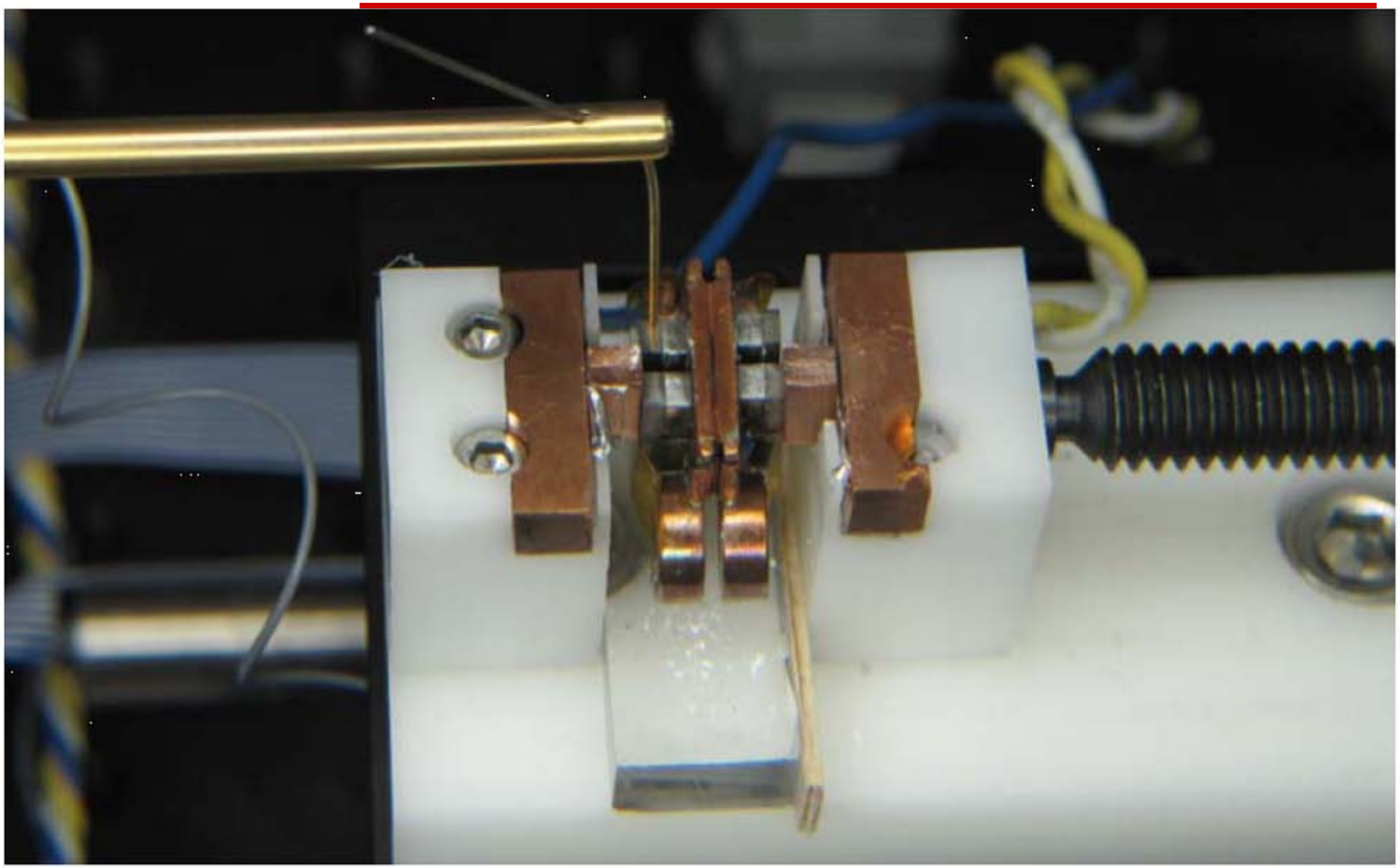
4.2 Electrical contact resistances
ZTPlus

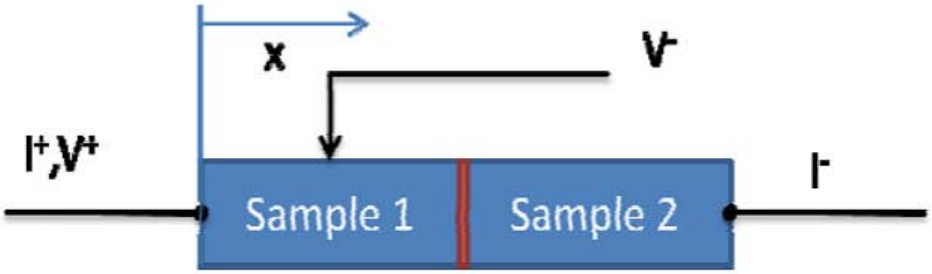
4.3 Thermal contact resistances (new technique)
Virginia Tech

4.4 Device performance
BSST

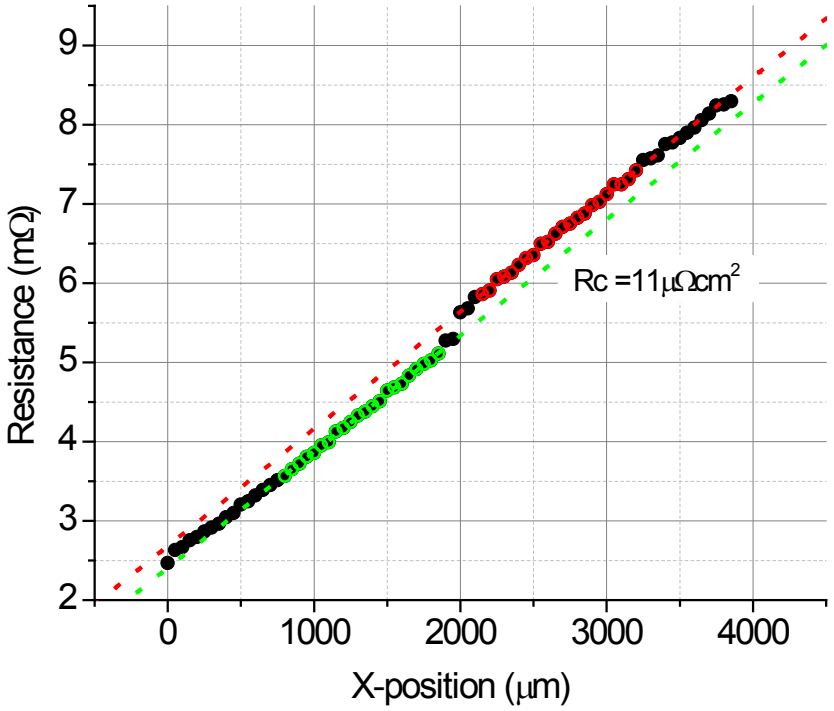
4.5 Circular cross-check is now possible
=> determination of the error bars

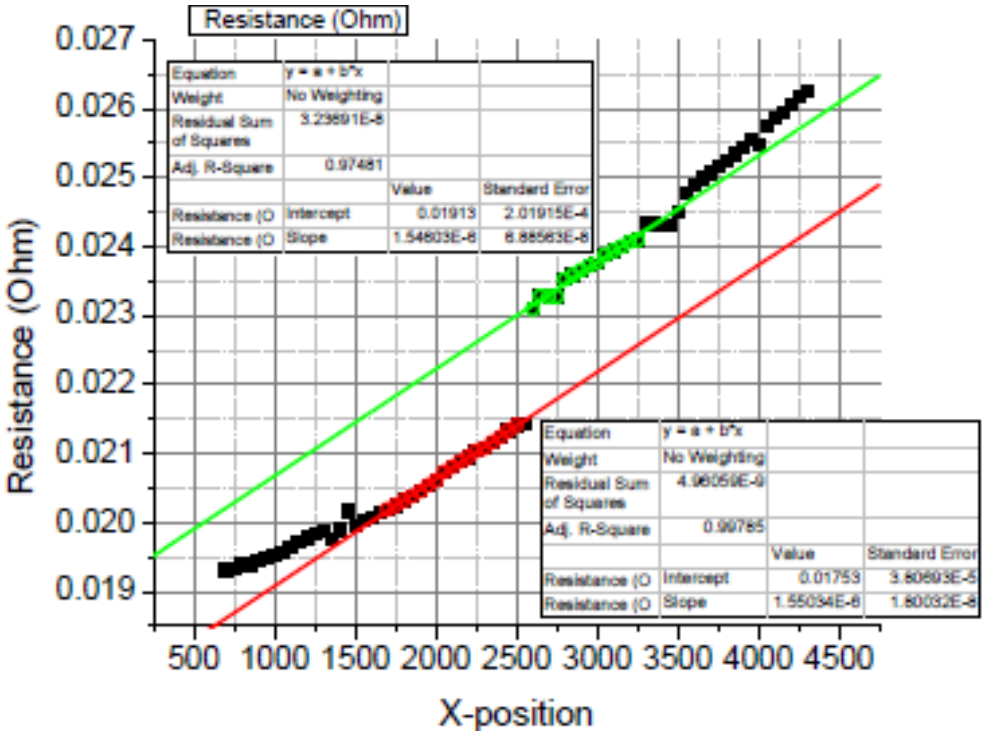
4.1 Electrical contact resistances



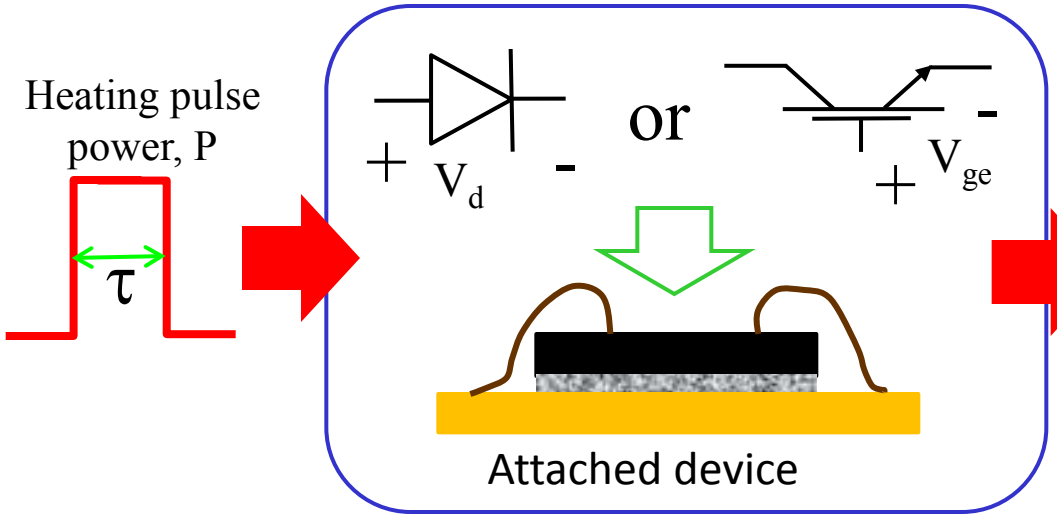


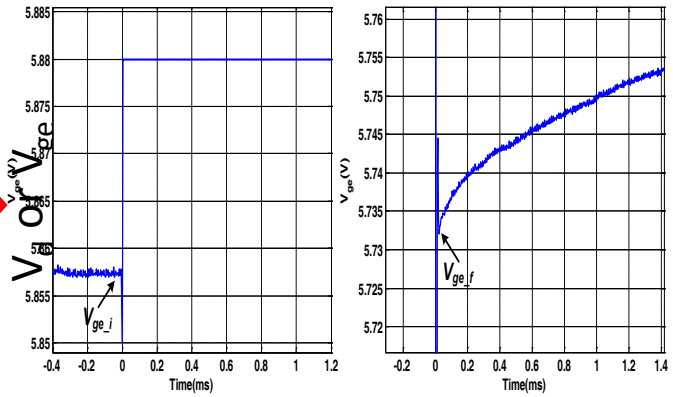
4.1 Electrical contact resistances





Thermal impedance characterization of the sintered joints

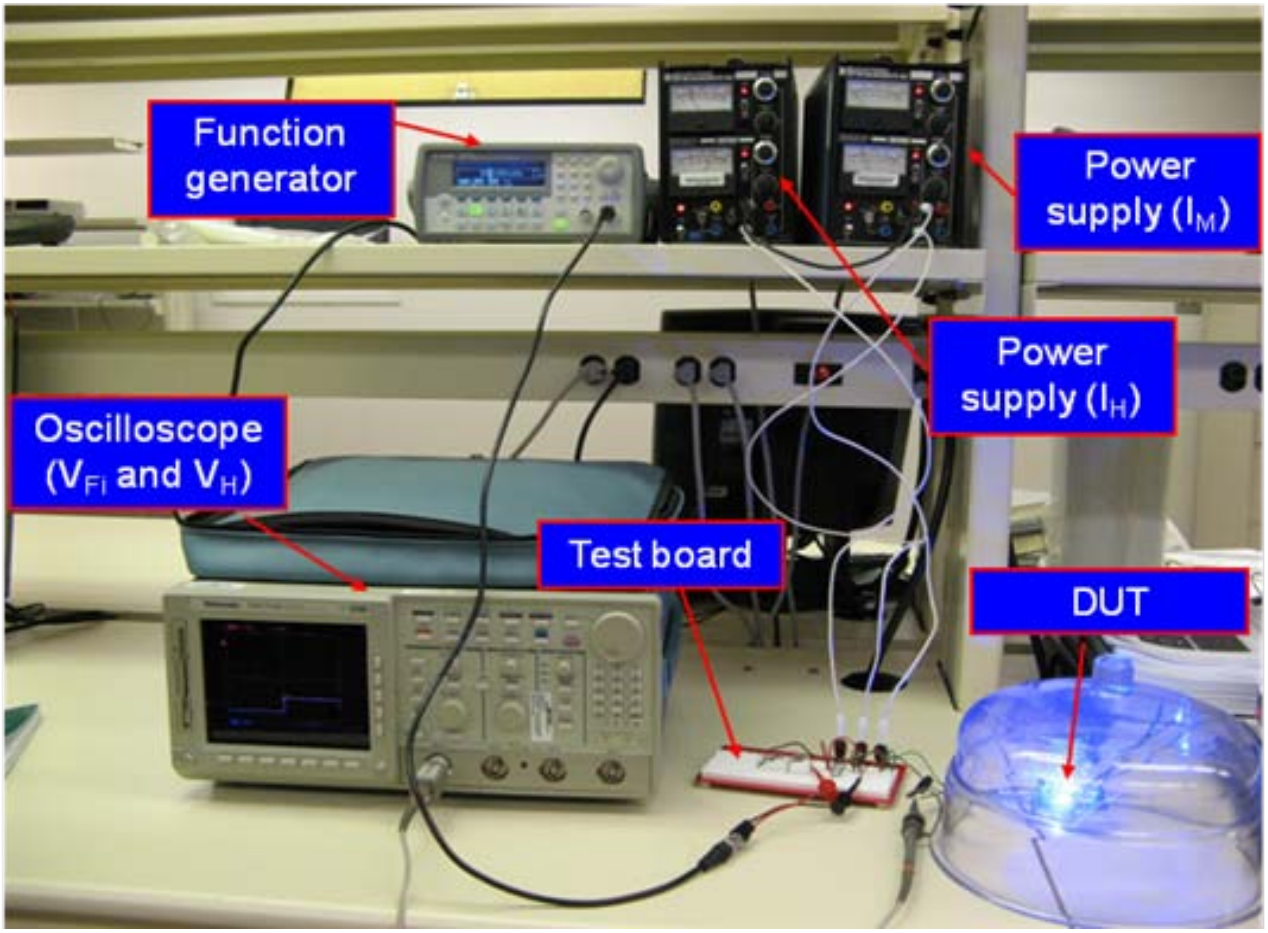




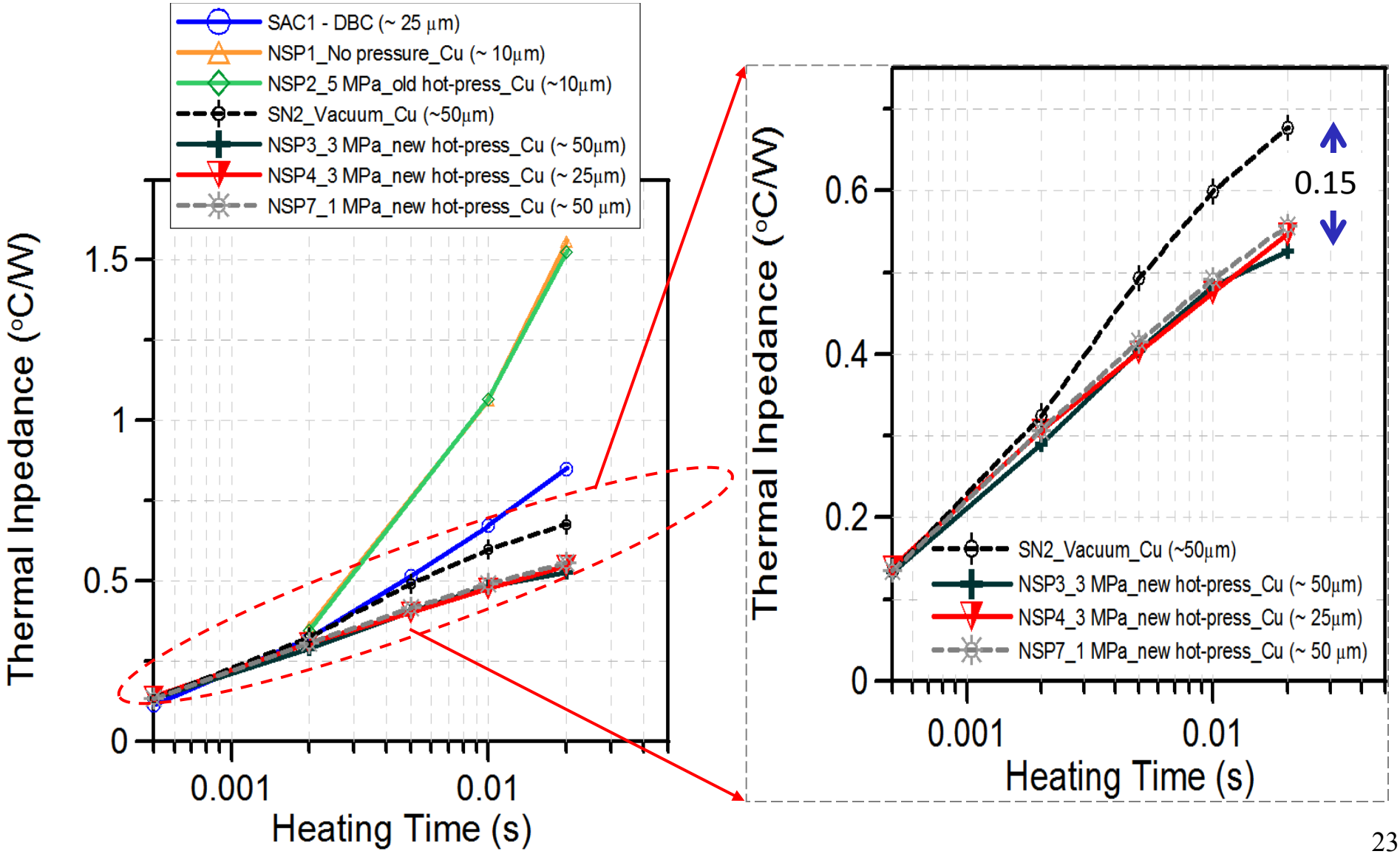
$$\text{Thermal impedance: } Z_{th} = \frac{\Delta T}{P_H} = \frac{\frac{\Delta V}{K}}{I_H V_H (1-\varphi)} = \frac{V_{Fi} - V_{Ff}}{I_H V_H (1-\varphi) K}$$

V_{Fi}: The initial VF value before application of heating power;
V_{Ff}: The final VF value after application of heating power;
I_H: The current applied to the DUT during the heating time in order to cause power dissipation;
V_H: The heating voltage resulting from the application of IH to the DUT;
P_H: The heating power pulse magnitude; product of VH and IH;
K: Thermal calibration factor, in mV/°C;

Setup for measuring thermal impedance, Z_{th}



Z_{th} results of a ~ 20-mm² IGBT attached by solder and nanosilver



5. Durability

1. Durability is built into every step of the design

2. Extensive durability testing at BSST and ZTPlus

BSST and ZTPlus, have state-of-the-art durability testing facilities used in the development of automotive products.

Ensure that this project incorporates automotive durability standards.

SUMMARY: FIVE THRUSTS

1. New Materials
    1. Two systems: PbSe and $Mg_2(Si, Sn)$
    2. Two techniques: nanostructuring by liquid encapsulation, resonant levels

2. New thermal design: cross-flow heat and current

3. New Interface technologies
    1. Compliant highly conductive contacts, Ag nanopaste
    2. In-situ metallization during pellet consolidation
    3. Deposition techniques

4. Metrology
    1. New thermal contact resistance measurement technqiue
    2. Self-consistency check: materials/interfaces/devices

5. Reliability
    1. Design-inherent
    2. Automotive-level durability testing