Superlattice Thermoelectric Materials and Devices

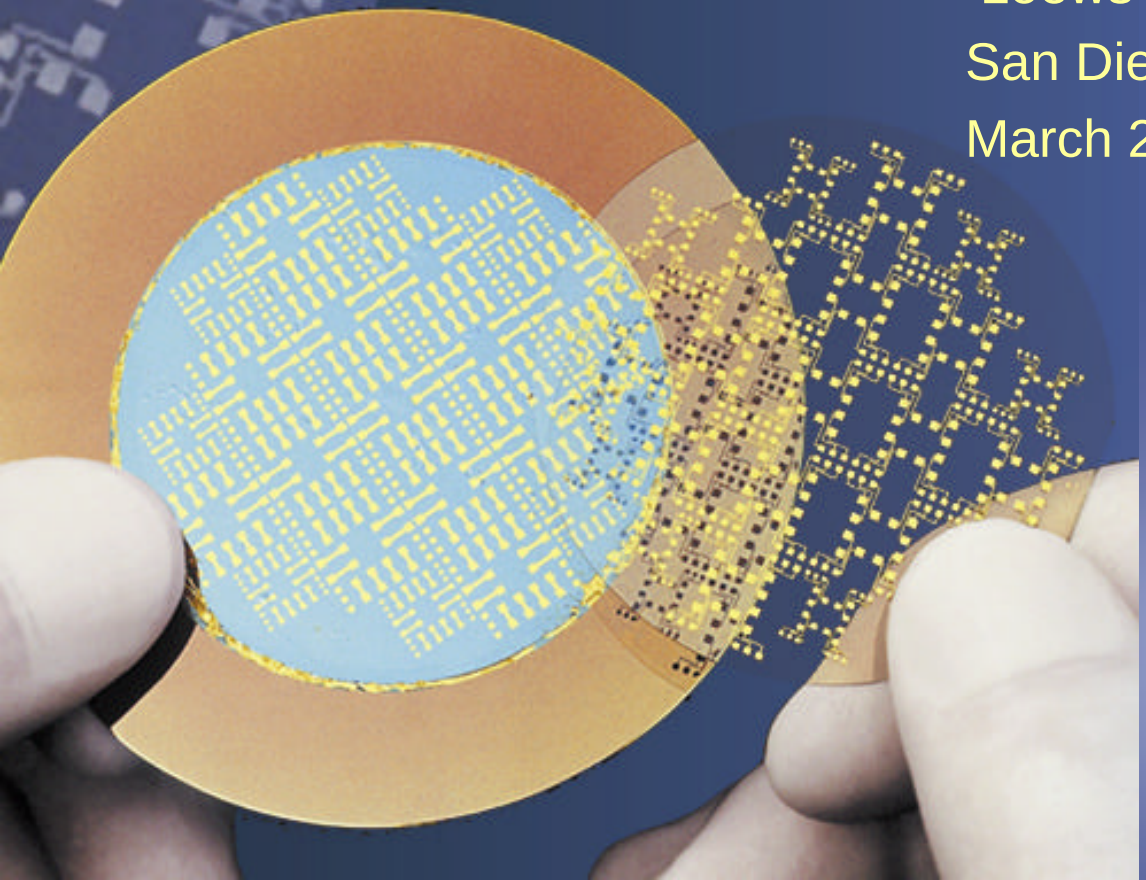
DARPA/ONR Program Review and

DOE High Efficiency Thermoelectrics Workshop

Loews Coronado Bay Resort

San Diego, CA

March 26, 2002



Dr. Rama Venkatasubramanian

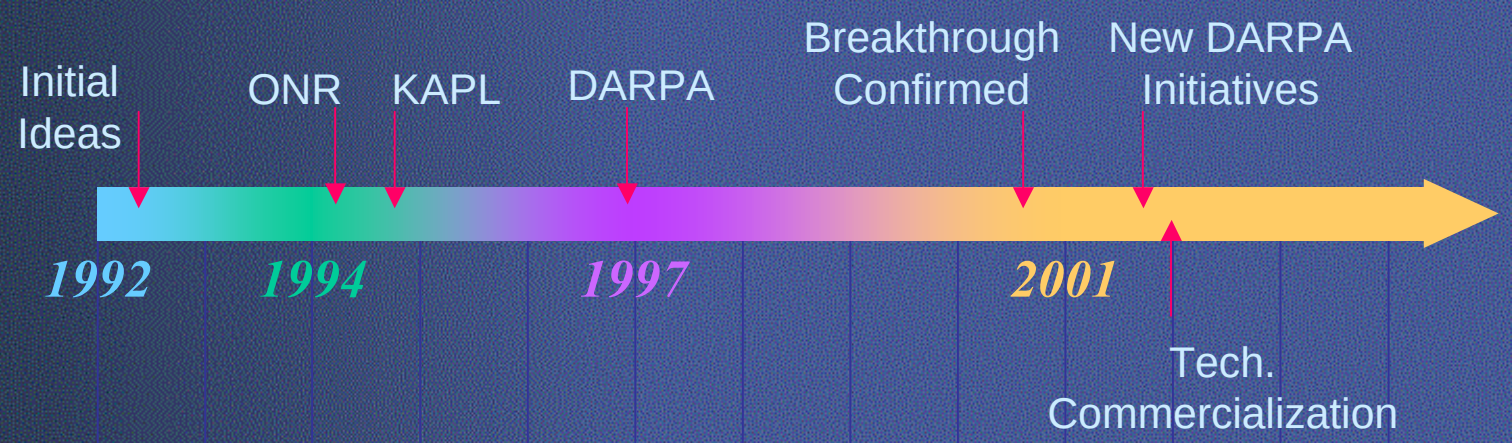
RTI INTERNATIONAL

RTI's Thermoelectrics R&D Team

Materials	Mr. Thomas Colpitts Mr. Brooks O'Quinn
Device and Module Development	Mr. Edward Siivola Mr. Kip Coonley Ms. Pratima Addepalli Mr. Michael Puchan
Thermal Management and Heat- Sources	Dr. John Posthill Mr. Robert Hendry
System Integration and Applications Development	Mr. Randall Alley Mr. Michael Mantini Mr. Dean Brooks

Acknowledgements

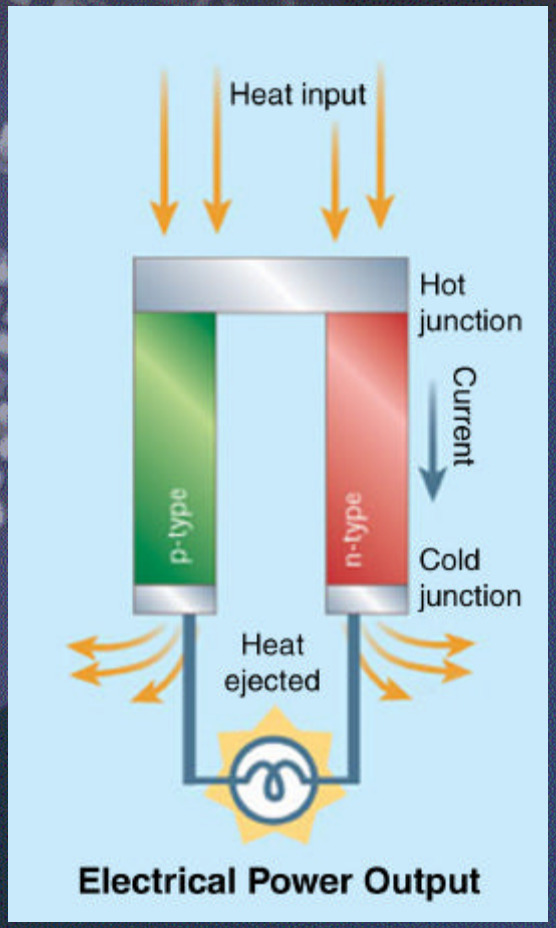
- ONR
- KAPL (DOE)
- DARPA
- RTI Technology Commercialization Fund and Technology Development Partners

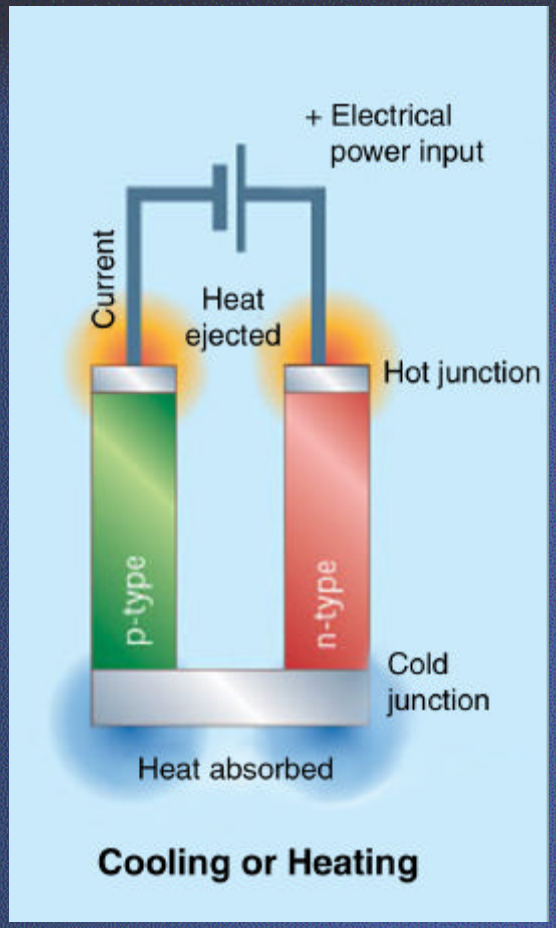


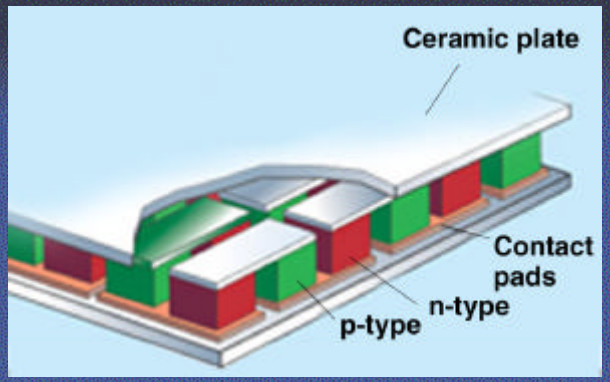
Outline

- Background to RTI's Breakthrough Thermoelectric Technology
- RTI's Progress in Cooling Module Development (Dr. Browning, Dr. Pazik, Dr. Wahl))
- Palm-Power Conversion Devices (Dr. Nowak)
- Projections for RTI's Superlattice TE Technology and Emerging Applications

Thermoelectric Effect







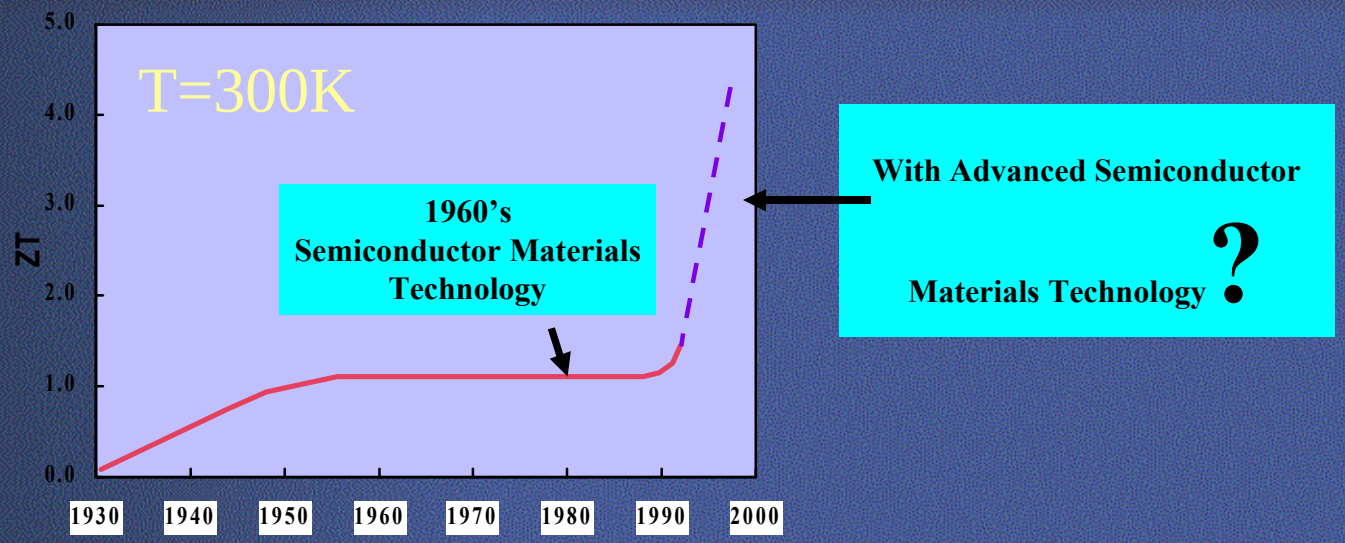
- Cooling or Power Conversion Efficiency critically dependent on the material Figure of Merit (ZT)
- $ZT = (\alpha^2\sigma/K)T$
- Minimize thermal conductivity and maximize electrical conductivity

Ref :Vining, Nature, 413, 577 (2001)

Why Thermoelectrics?

- Converts electrical and thermal energy using a solid state device with minimal moving parts
- If efficient, opens up a wide array of potential uses
- Chip-scale functionality with thin-film devices using standard microelectronic processing
  - Computer Chip, Photonic Chip, Lab-on-a-Chip

THERMOELECTRIC TECHNOLOGIES in 1992
Fort Belvoir Workshop Organized by Dr. Stuart Horn



- ZT - figure of merit - has been stagnant at about 0.8 to 1 for the last 40 years - cooling and power conversion efficiencies low - useful only in applications where you <u>really need</u> it
  - More importantly, cooling power density (P_D) in bulk devices are low to be of any interest for many electronics cooling applications
- ZT need to improve over 1.5 at 300K for a major impact and around 3 for a revolutionary impact; larger P_D would also open up new applications

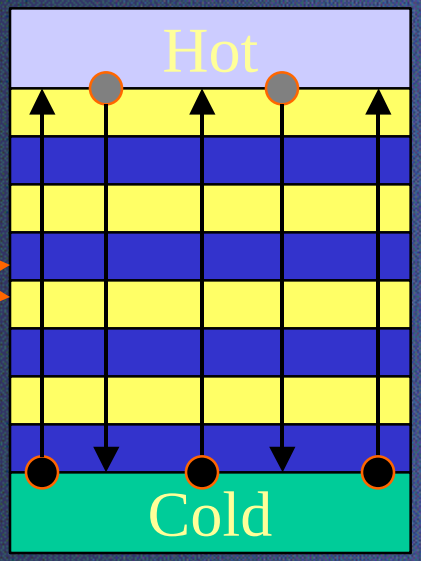
Since then what are some of the approaches for ZT Enhancement ?

- **Bulk Materials**
  - Filled Skutterudites (Slack, JPL, Sales et al.) - Have Atoms that "Rattle" in the large unit cells to effectively scatter low-frequency, heat-conducting phonons
  - New Versions of Bi_2Te_3 such as $CsBi_4Te_6$ (Kanatzidis et al.) - Appears to be band-gap modification leading to a more optimized ZT at lower temperatures
- **Thin- Film Materials**
  - Quantum wells, Quantum Wires, Quantum Dots (Dresselhaus et al., Harman, Broido et al., Mahan et al.)
  - Superlattices for thermal conductivity reduction (Venkatasubramanian, Mahan, Chen)
  - Superlattices for improved electronic/hole transport from reduction of alloy scattering of carriers (Venkatasubramanian)
  - Thermionics in Heterostructures (Mahan, Shakouri)

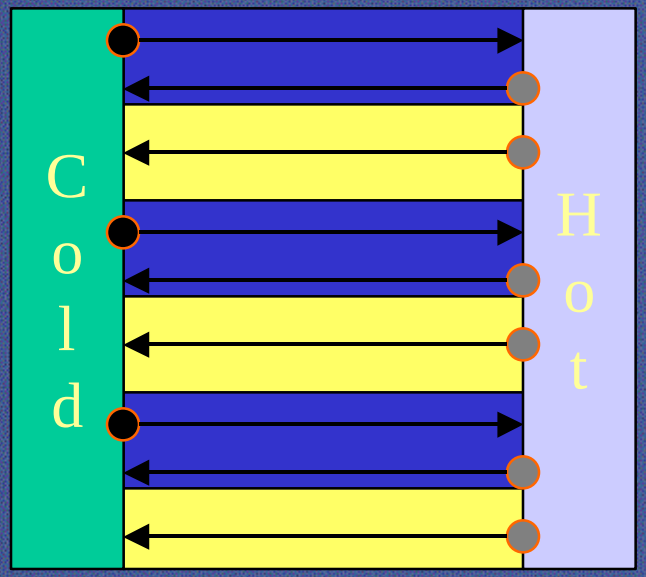
Comparison of Superlattice and Quantum-Well Approaches

Utilize interfaces to impede phonons that cause undesired reverse flow of heat but not impede forward-flowing electrons

Superlattice (RTI)



Quantum Well (MIT)



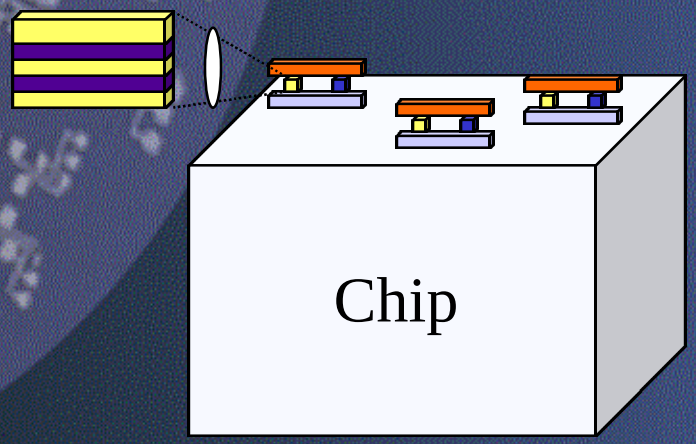
Utilize quantum confinement to get efficient heat energy transport by electrons

- Current and Heat Transport Perpendicular to the superlattice interfaces
- Easy Adaptability for Device Implemenation

- Current and Heat Transport Parallel to the Quantum-well interfaces

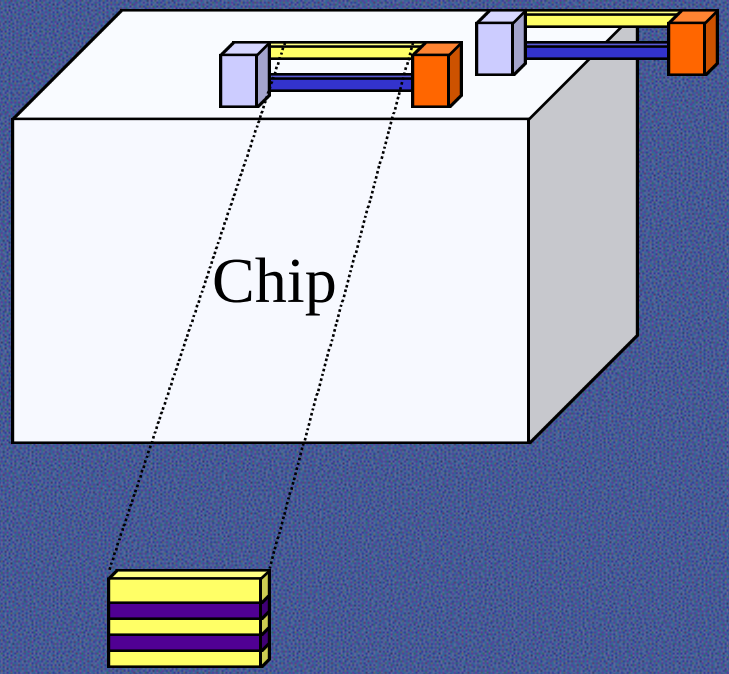
Comparison of Implementation of Cross-Plane and In-Plane Approaches in Devices

Cross-plane Low-Dimensional (Superlattice)



- Need Much Thinner Epi Layers
- Integrated Conventional Microelectronic Processing

In-plane Low-Dimensional (Quantum Well)



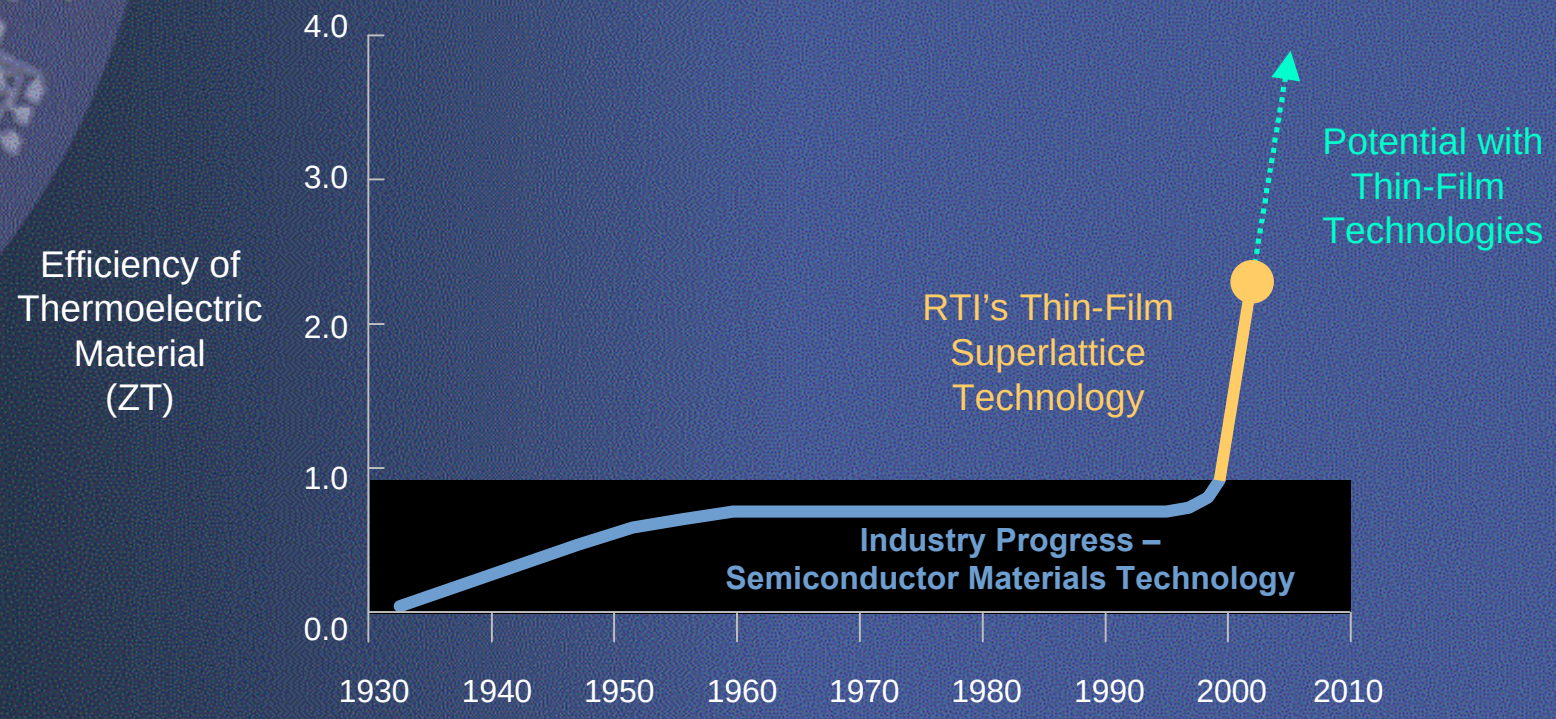
- Need Thick epi layers -expensive
- Need Bulk-like or MEMS-like Processing

Some of the Bulk Material and Thin-film Developments

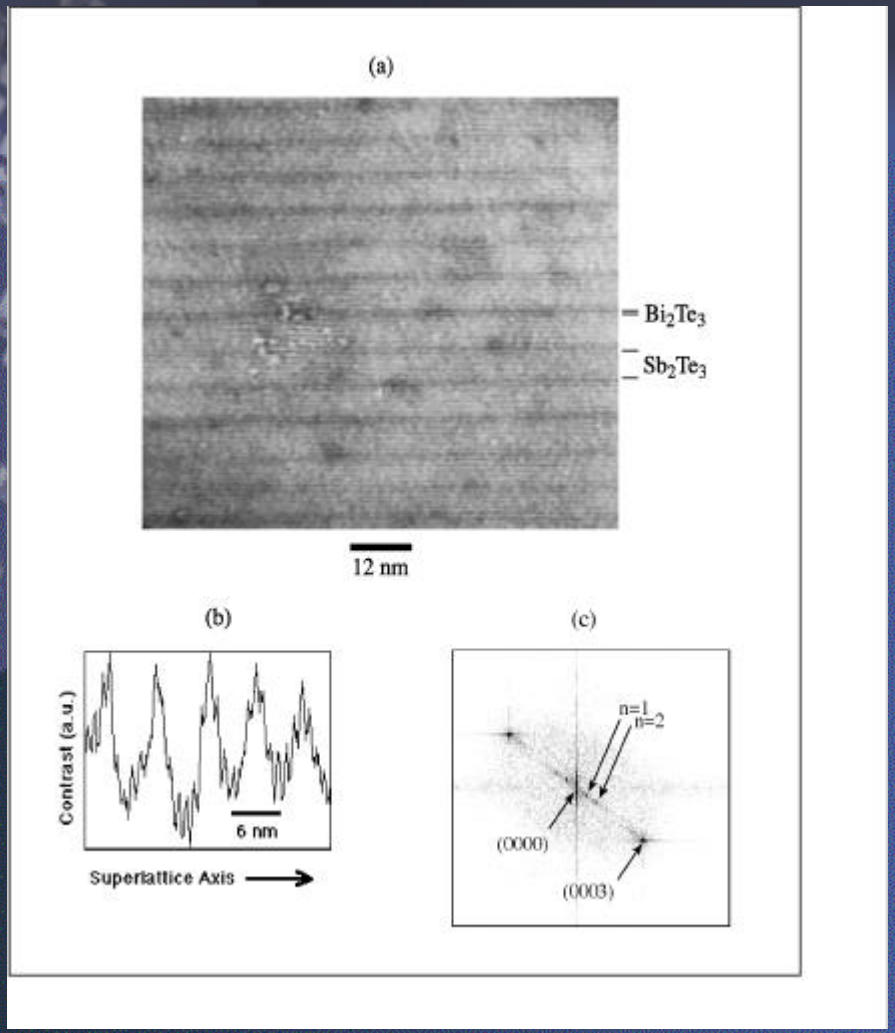
- $Cs\ Bi_4Te_6$ (Kanatzidis, Michigan State University)
  - Bulk Materials with a ZT~ 0.8 at 225K but less than 0.8 at 300K (*Science* **287**, 1024-1027, 2000)
- Filled Skuterrudites (Fleurial et al., JPL)
  - Bulk materials with a ZT ~1.35 at 900K (Proc. Of 15th International Conf. On Thermoelectrics, 1996)
- PbTe/PbTeSe Quantum-dots (Harman, MIT Lincoln Labs.)
  - ZT~2 at 550K and ZT~0.8 at 300K based on estimated thermal conductivity values (J. Electronic Materials, 29, L1 , 2000)
- Bi_2Te_3/Sb_2Te_3 Superlattices (Venkatasubramanian, RTI)
  - ZT~2.4 at 300K in devices with all properties measured at the same place, same time, with current flowing and verified by two independent techniques (*Nature*, 597-602, 2001)

RTI's 40-Year Breakthrough

- **$ZT_{material}$ > 2. 5 at 300K in p-type superlattices**
- **Device ZT ~2.4 at 300K through the development of ultra-low resistivity contacts**



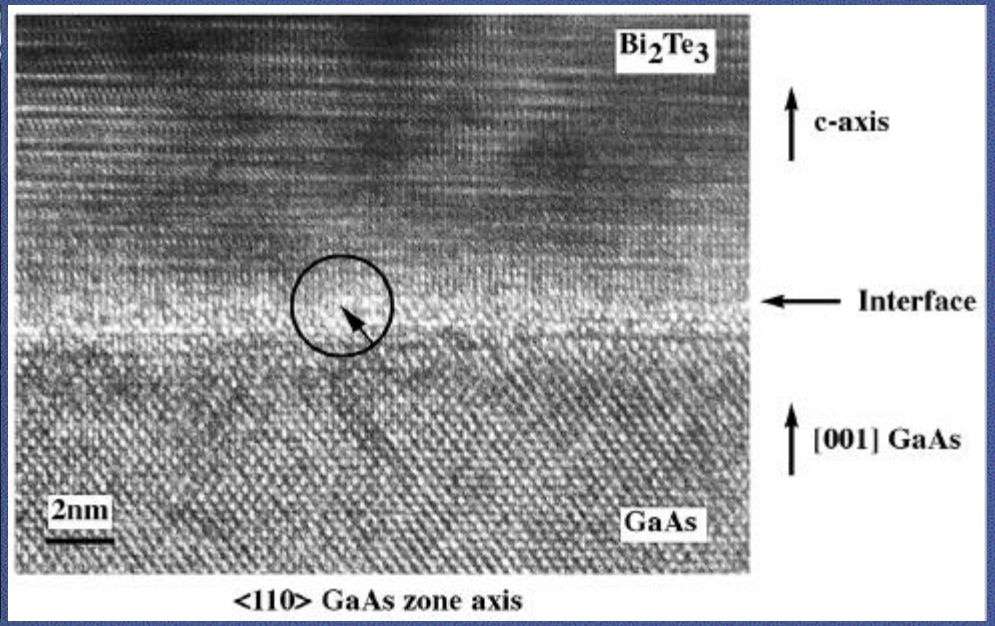
RTI's Superlattice Material



- 10Å/50Å Bi_2Te_3/Sb_2Te_3 Structure
- Optimized for disrupting heat transport while enhancing electron transport perpendicular to the superlattice interfaces

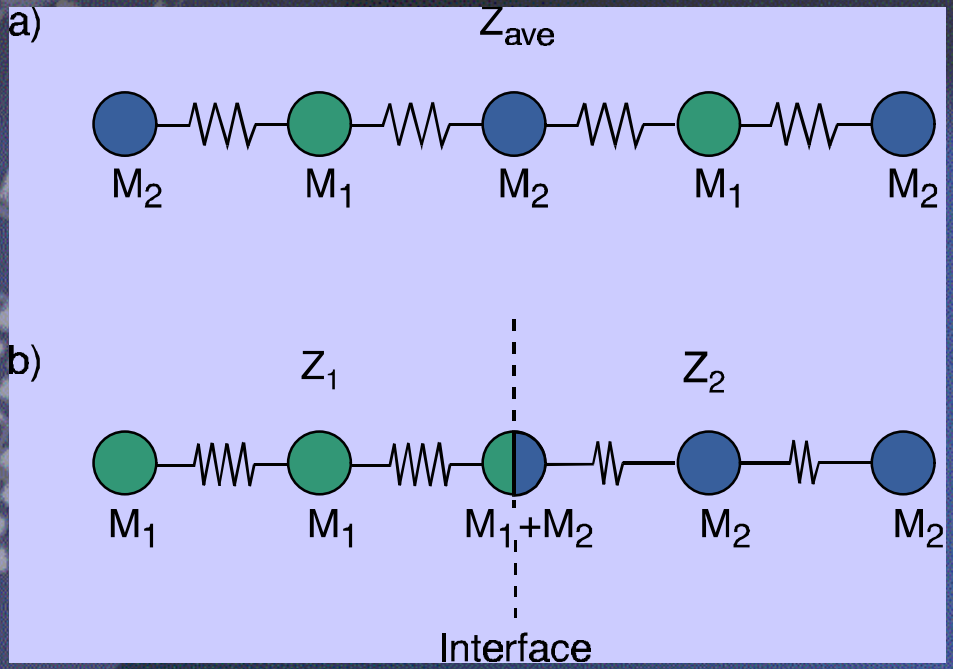
Applied Physics Letters, 75, 1104 (1999)

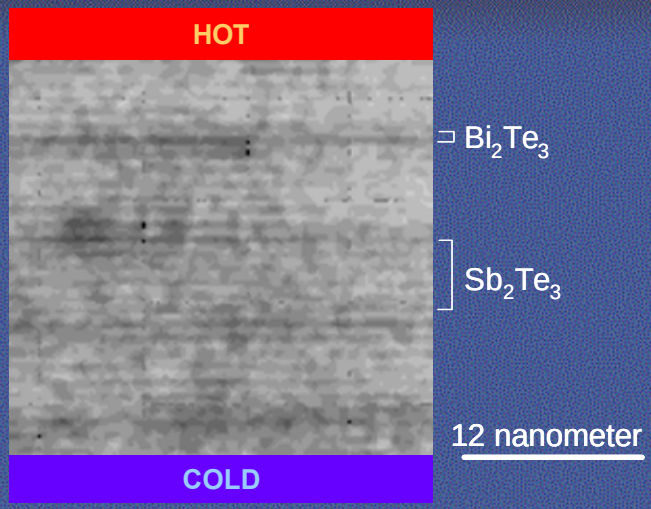
MOCVD Growth of Superlattices

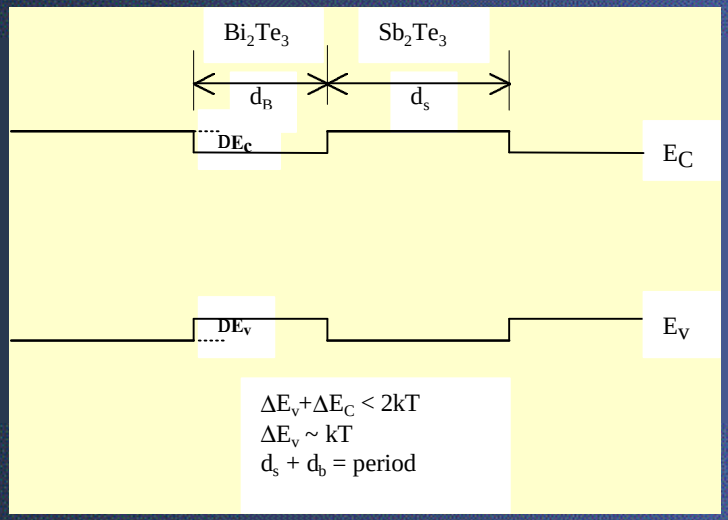


- Low-temperature technology
- In-situ ellipsometry for nanometer-scale control of deposition

Basis for the Superlattice Approach

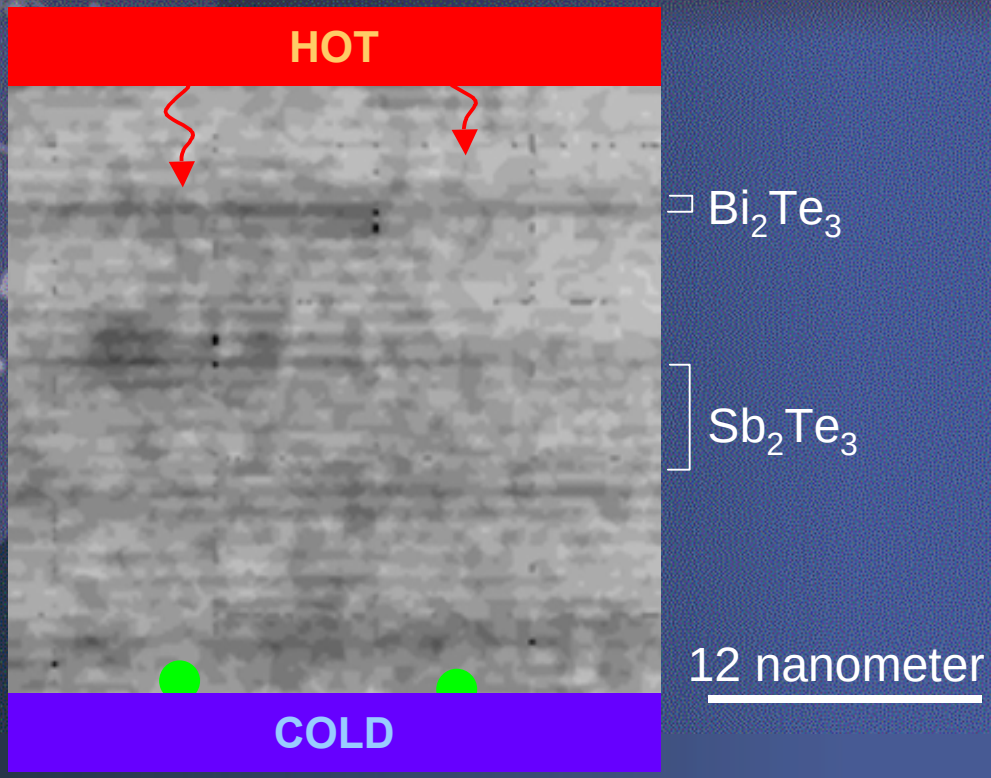






- The improvement arises from impeding phonon flow without disrupting electron flow
  - Phonon-Blocking, Electron Transmission

RTI's Superlattice Material



The RTI breakthrough arises from reducing heat transmission without disrupting electron flow

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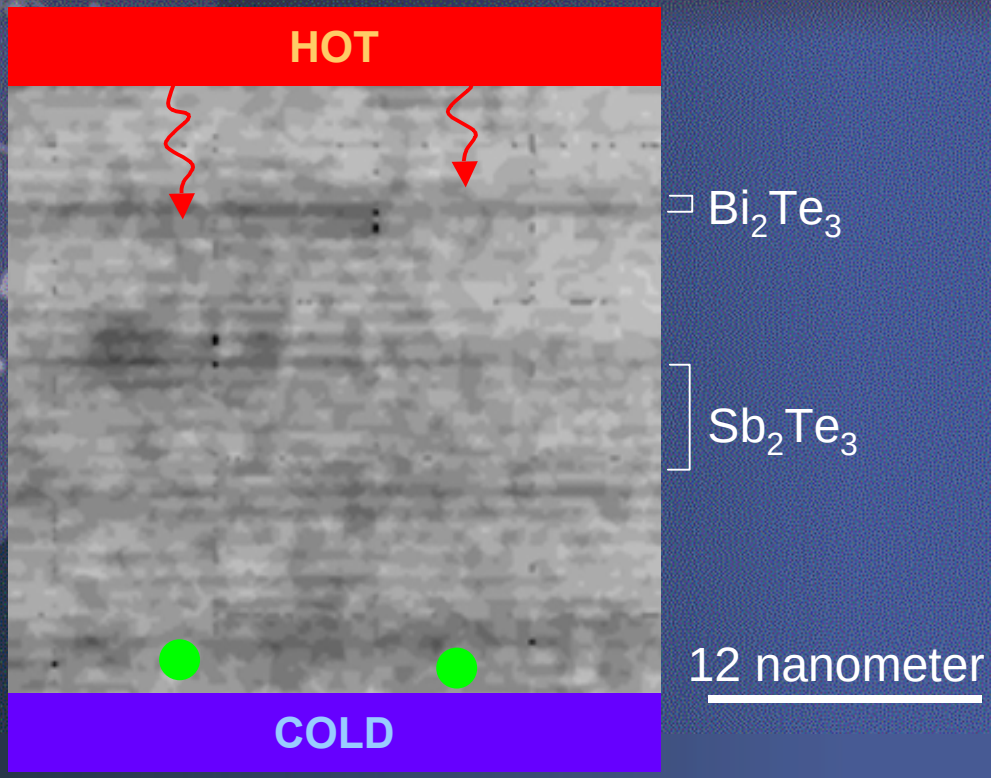

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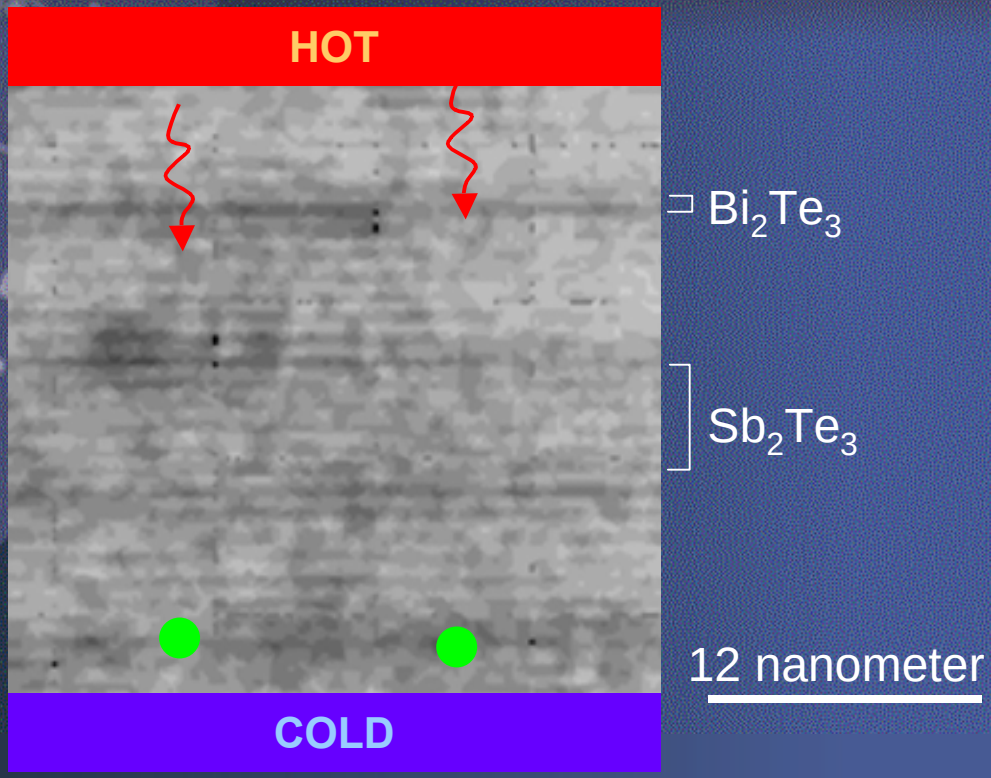

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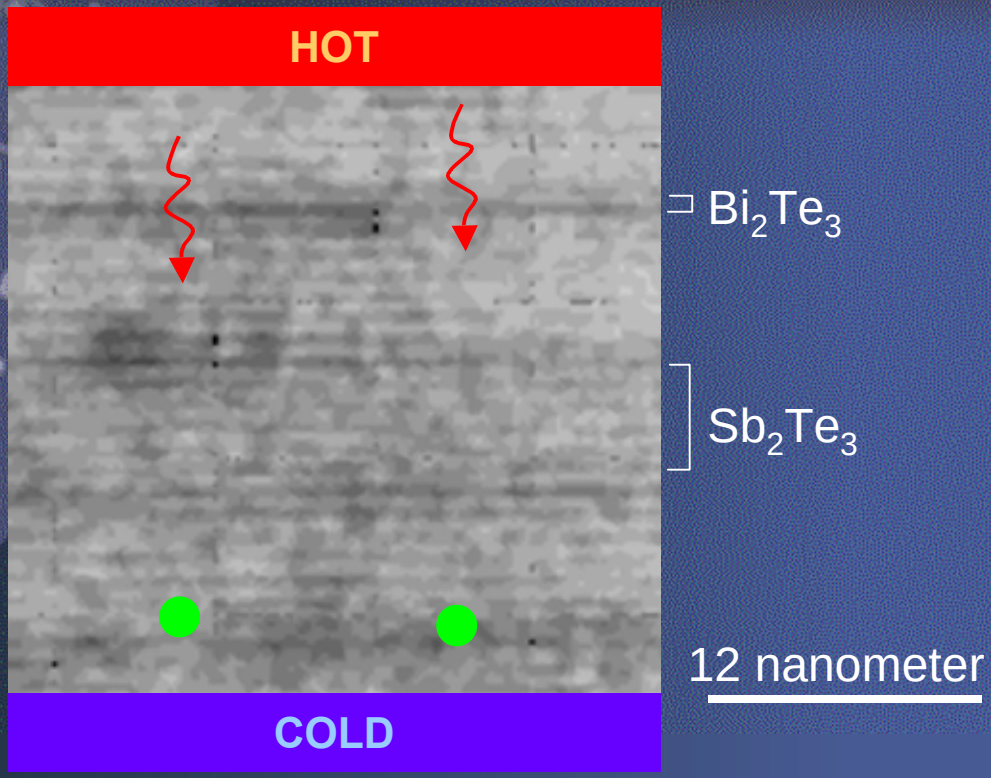

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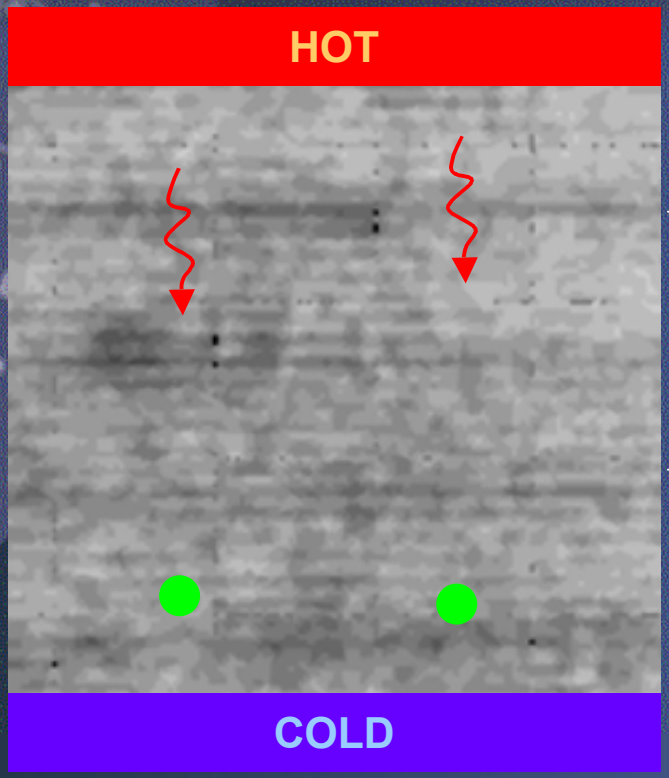

Bi_2Te_3

Sb_2Te_3

12 nanometer

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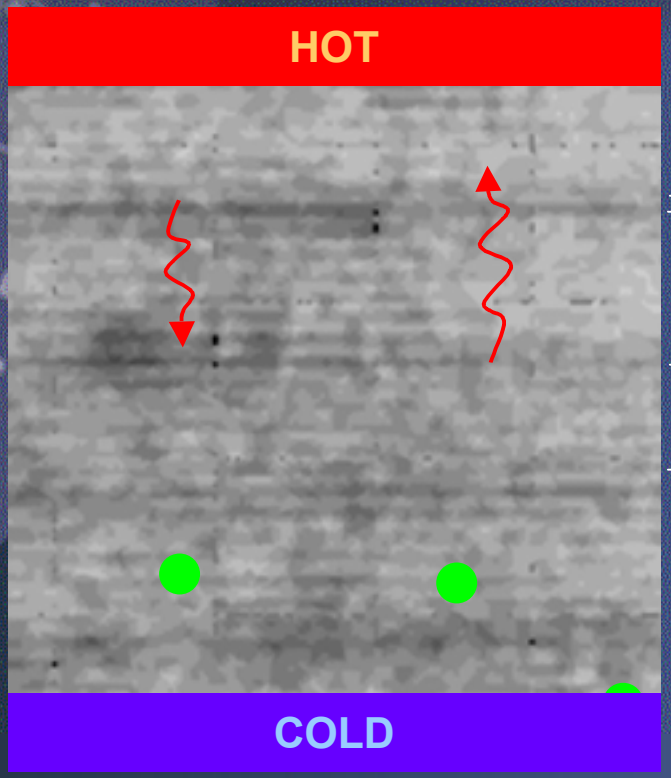

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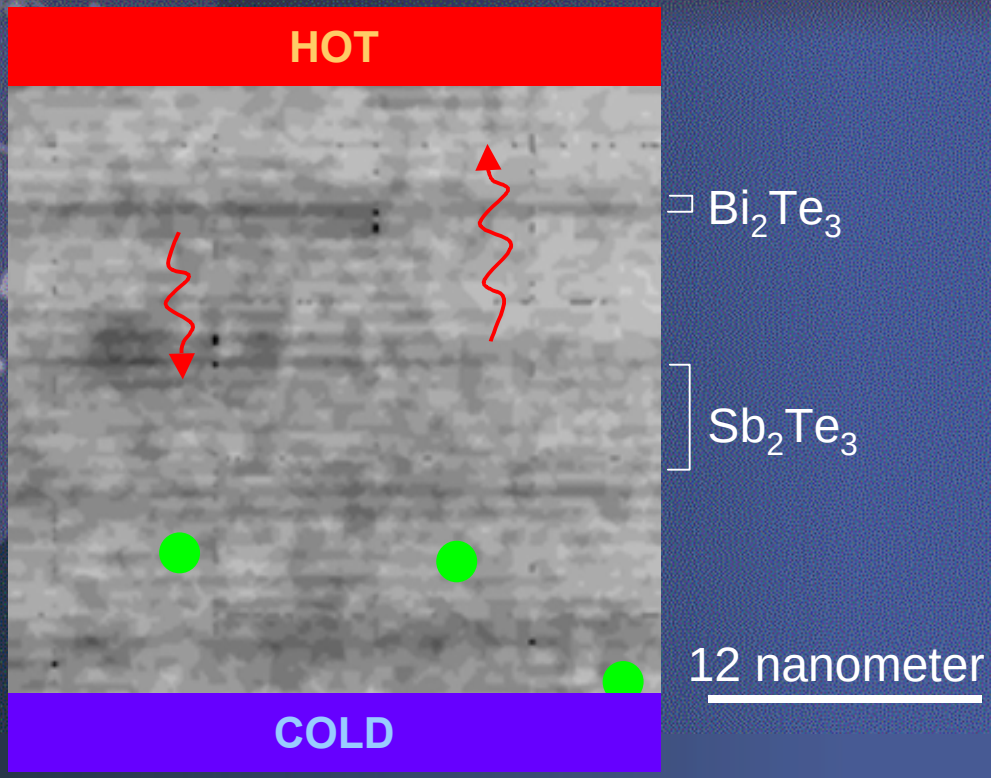

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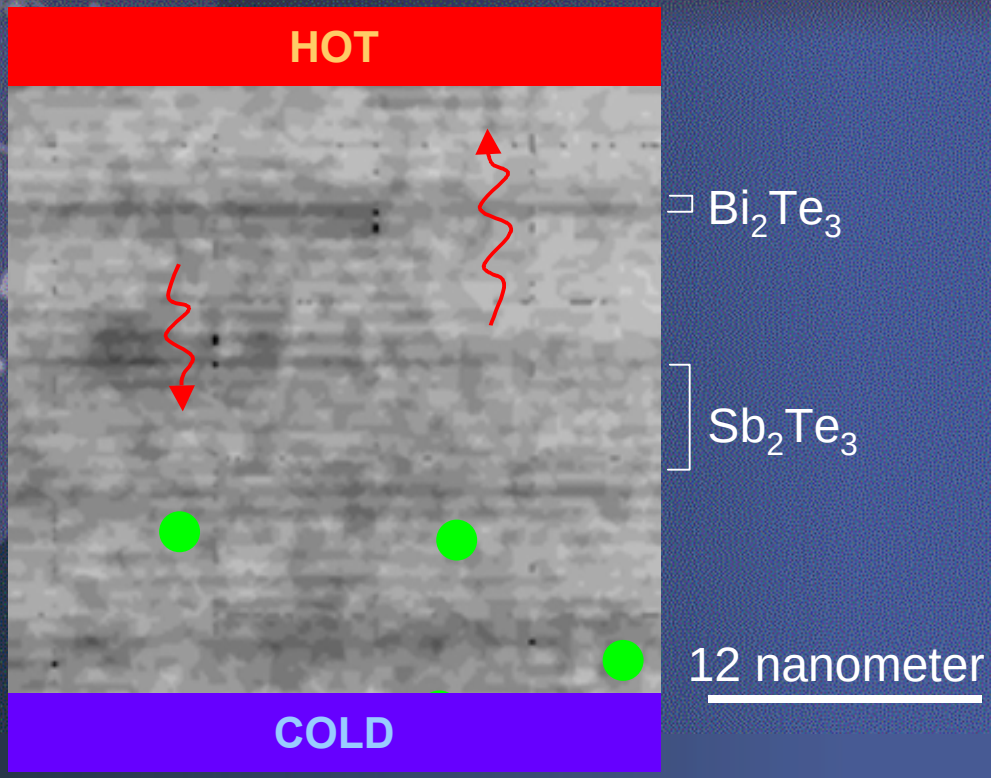

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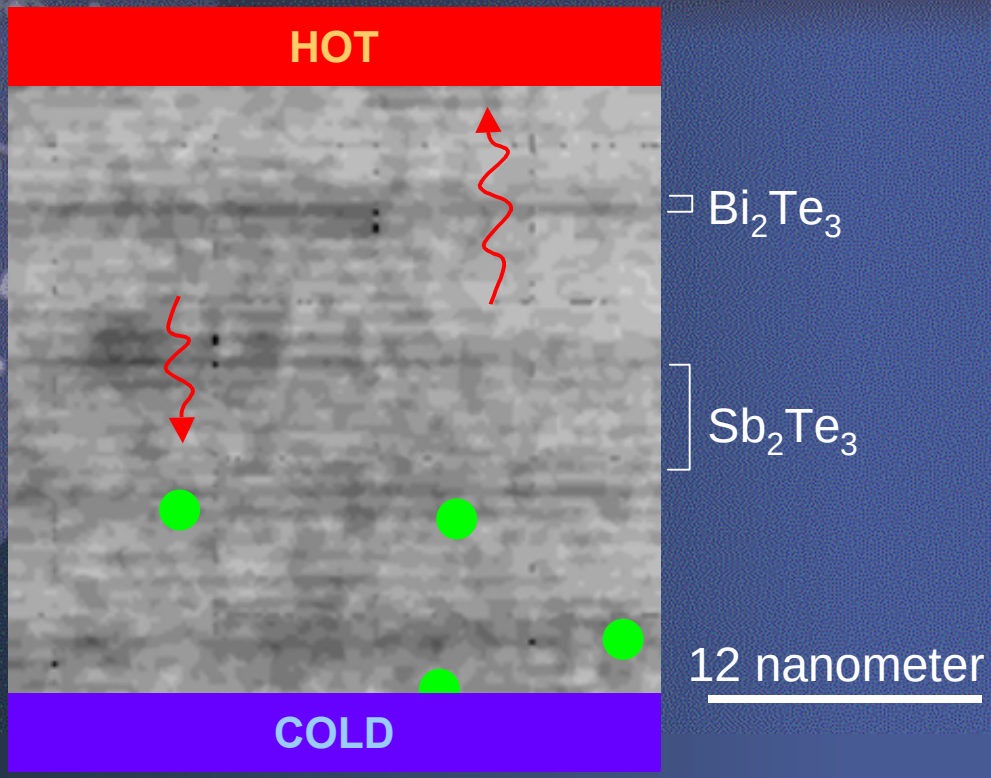

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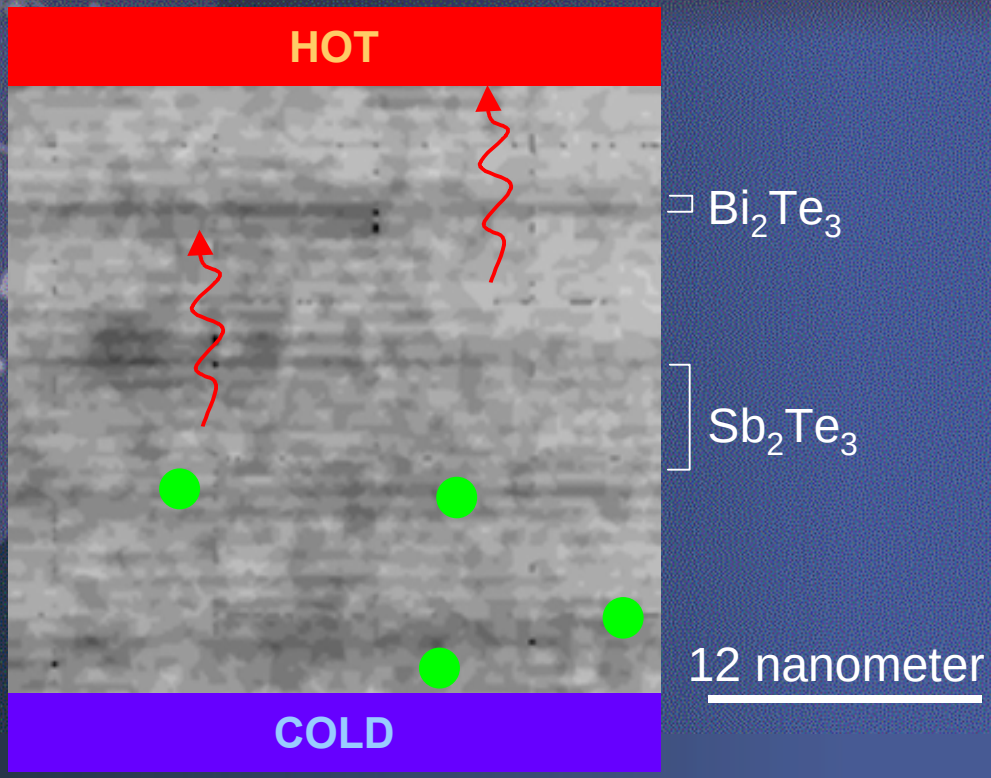

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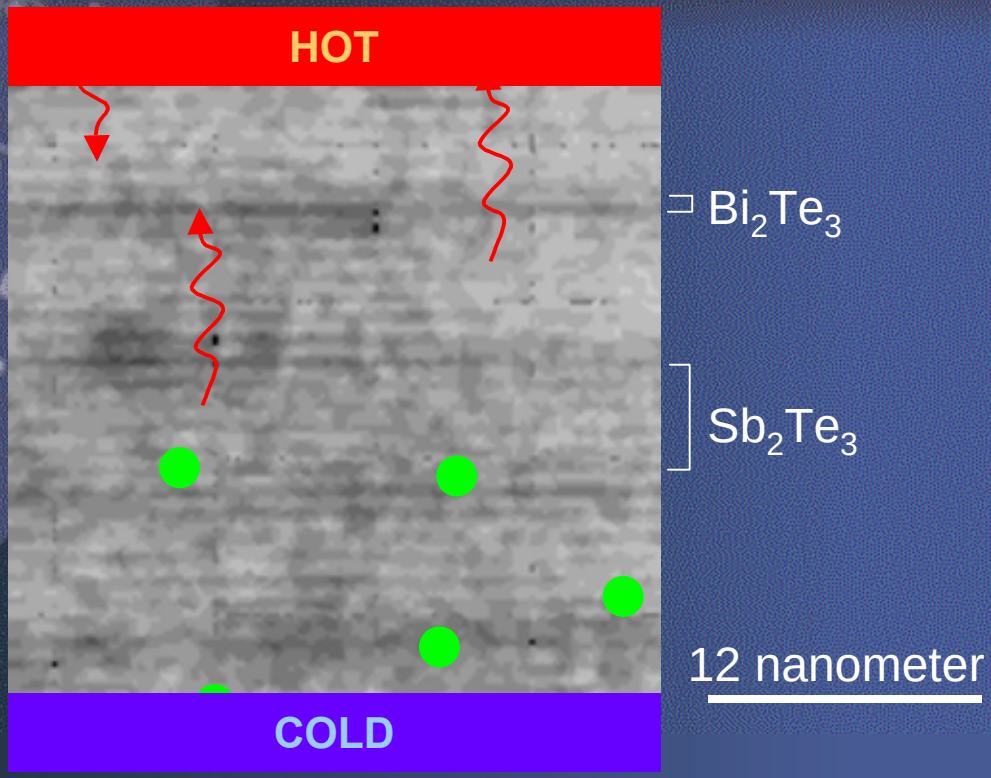

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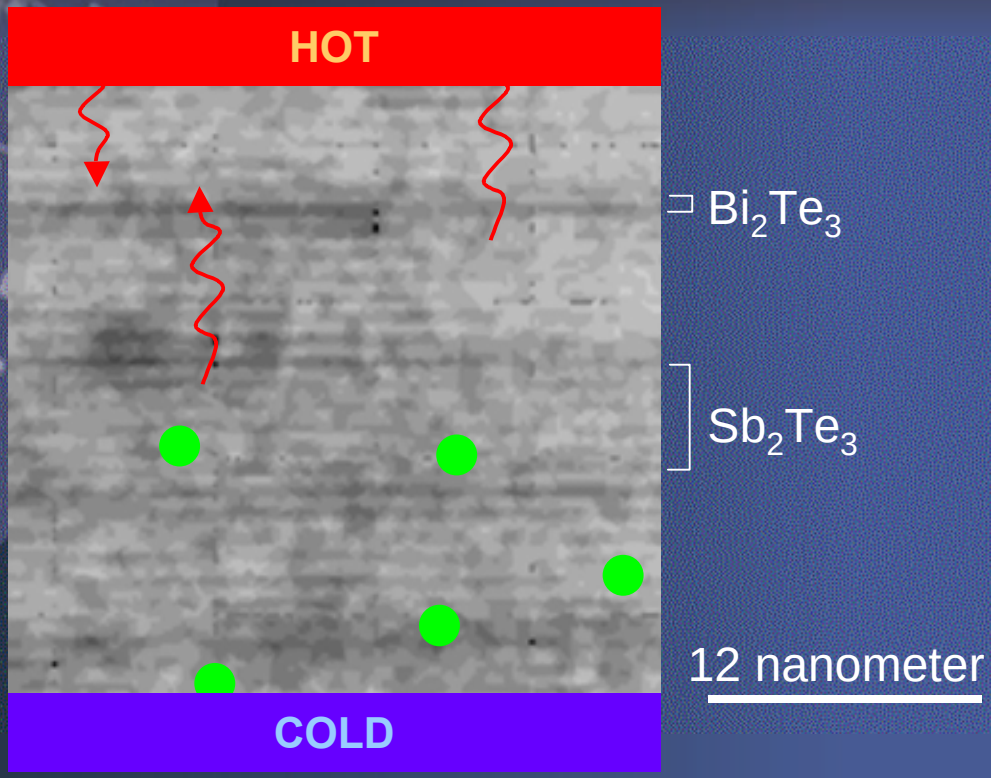

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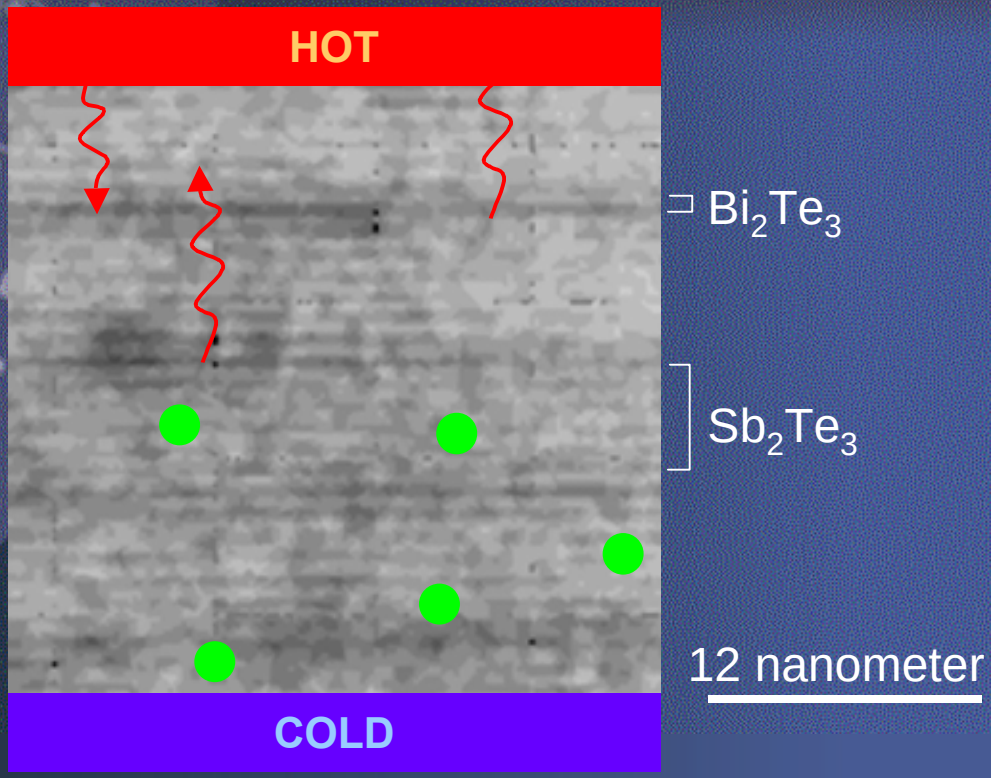

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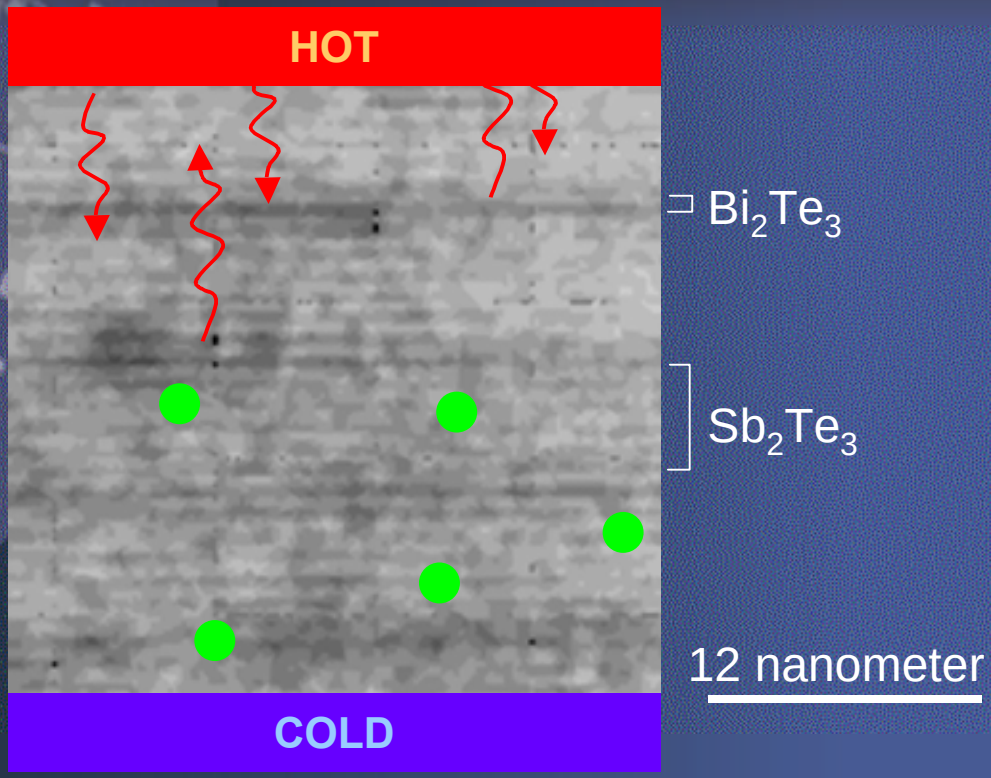

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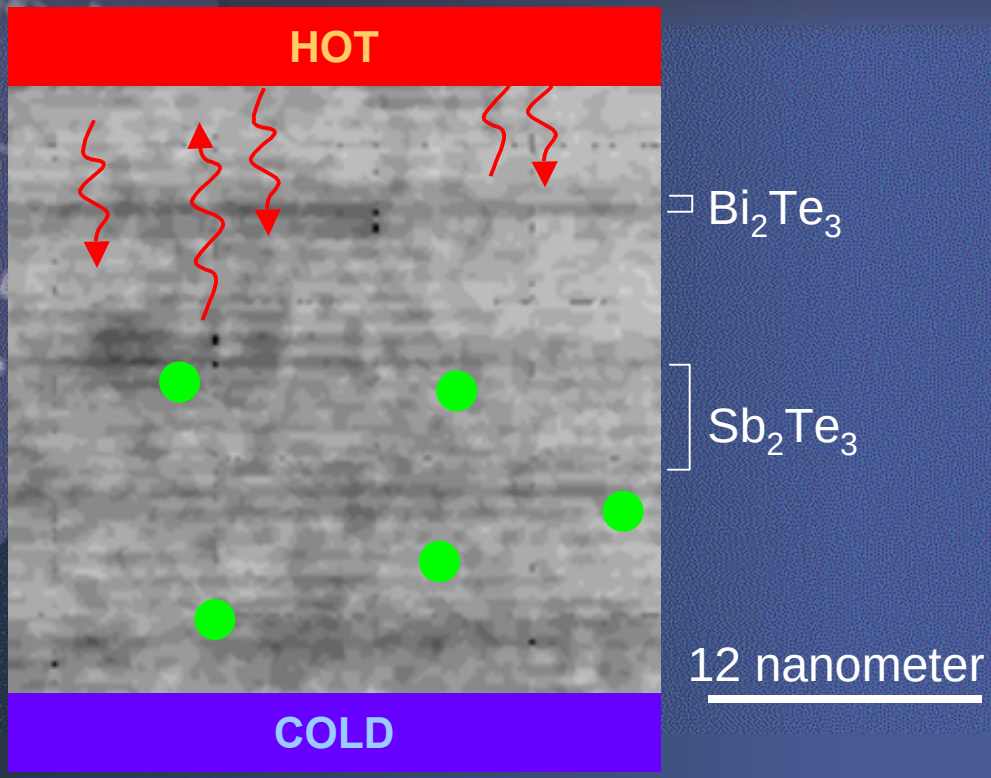

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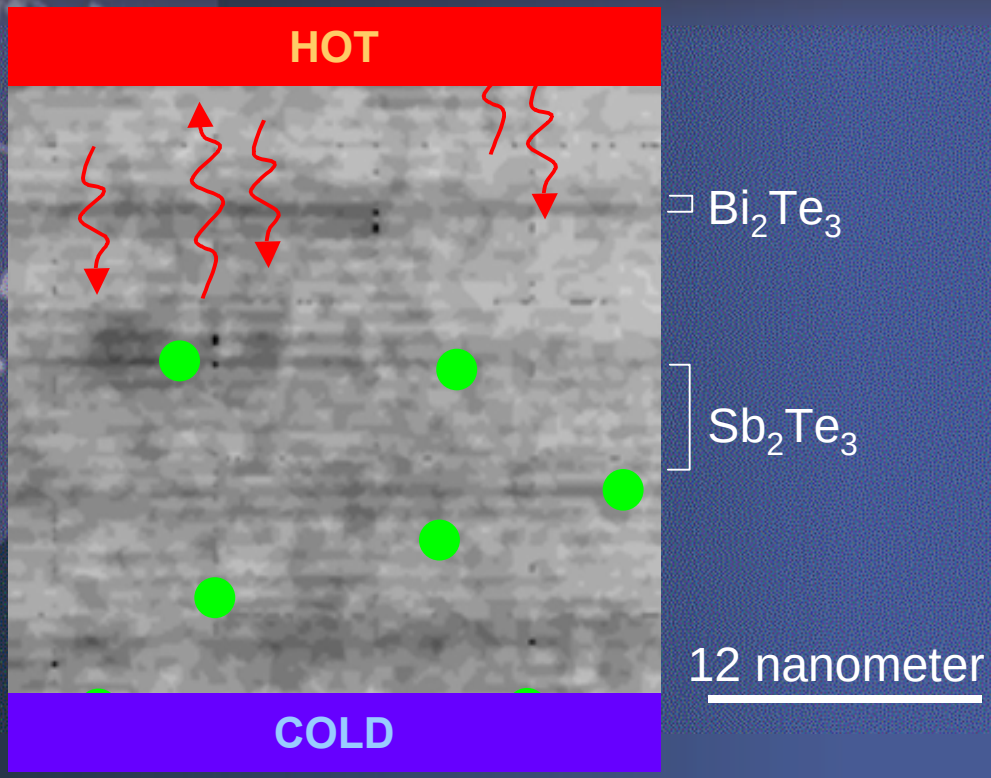

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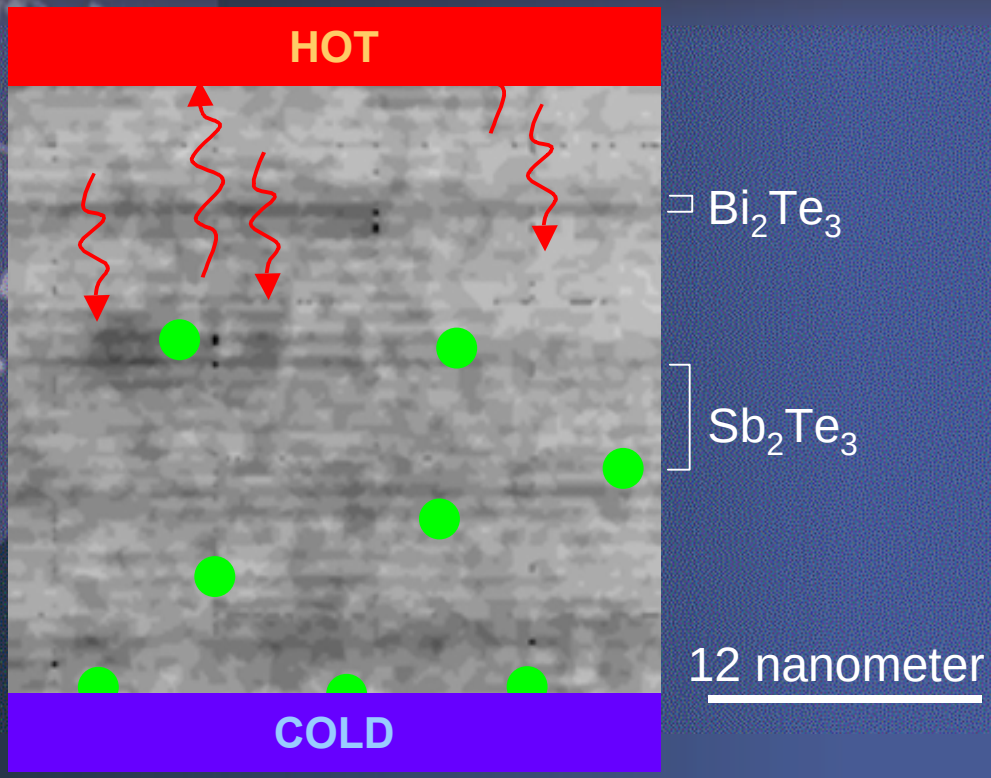

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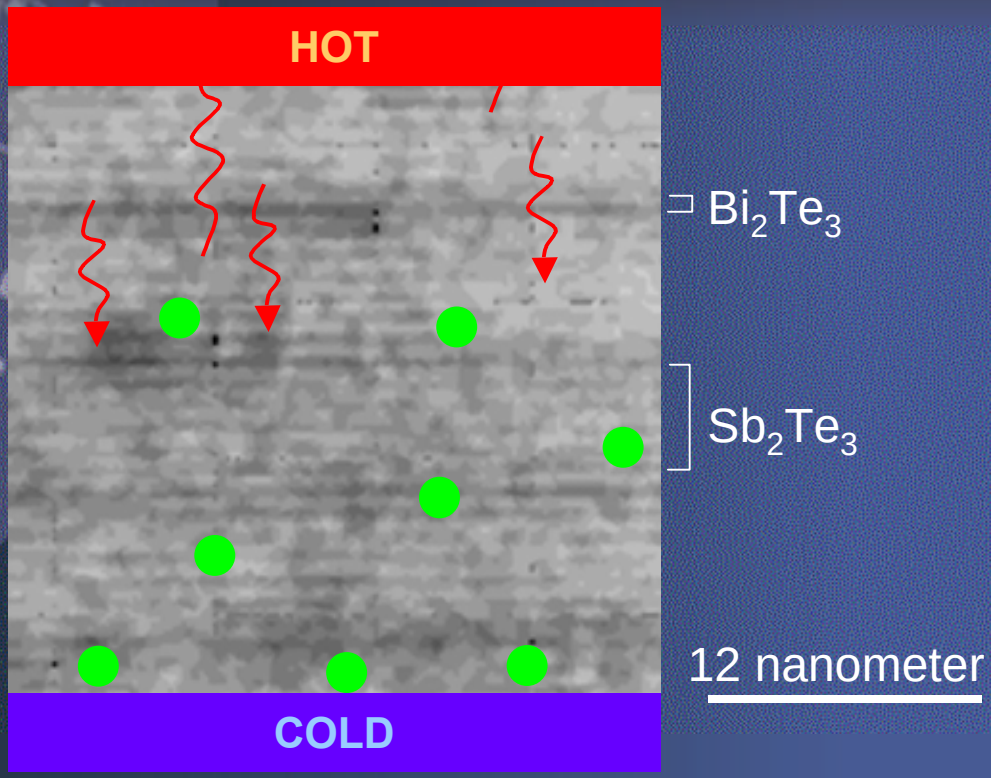

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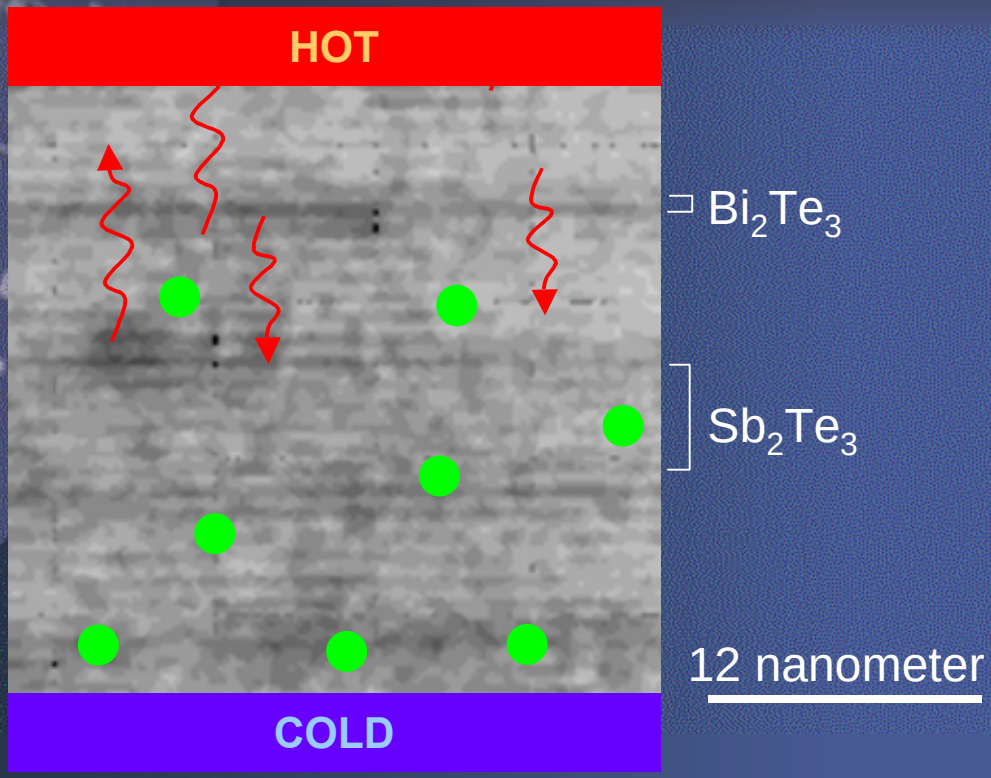

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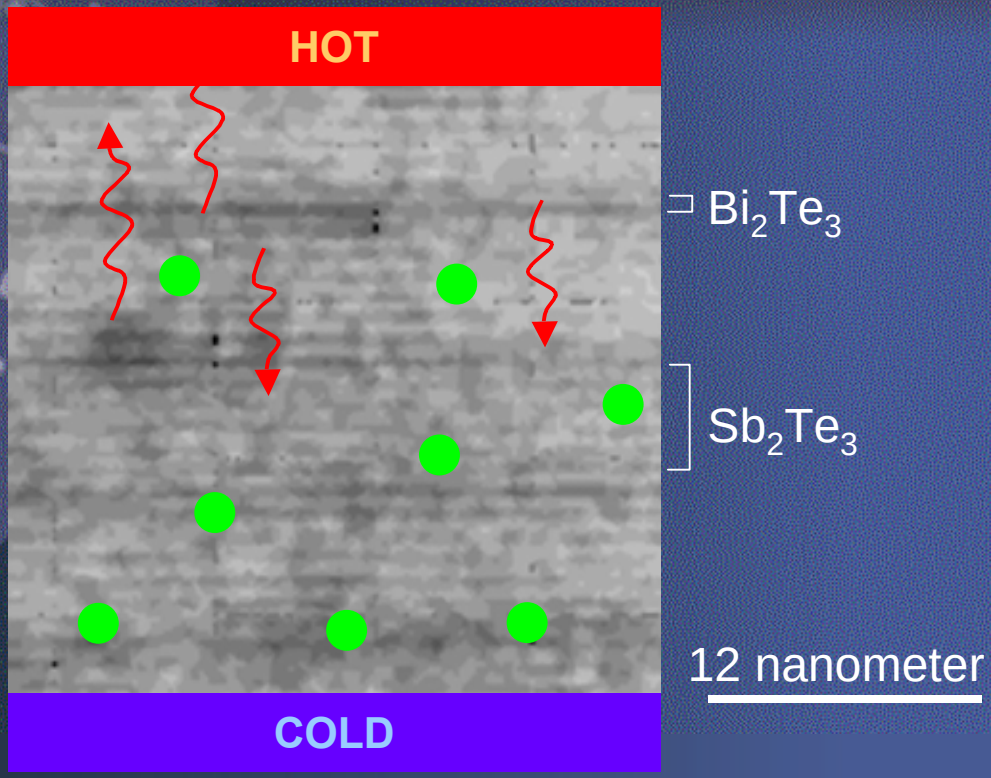

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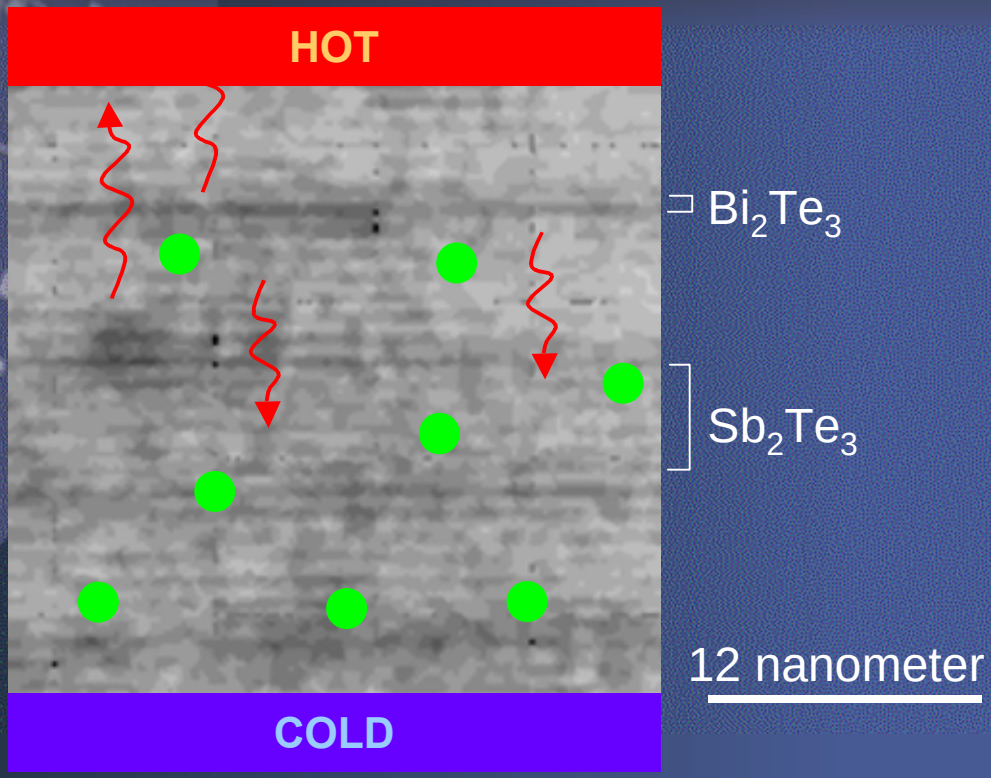

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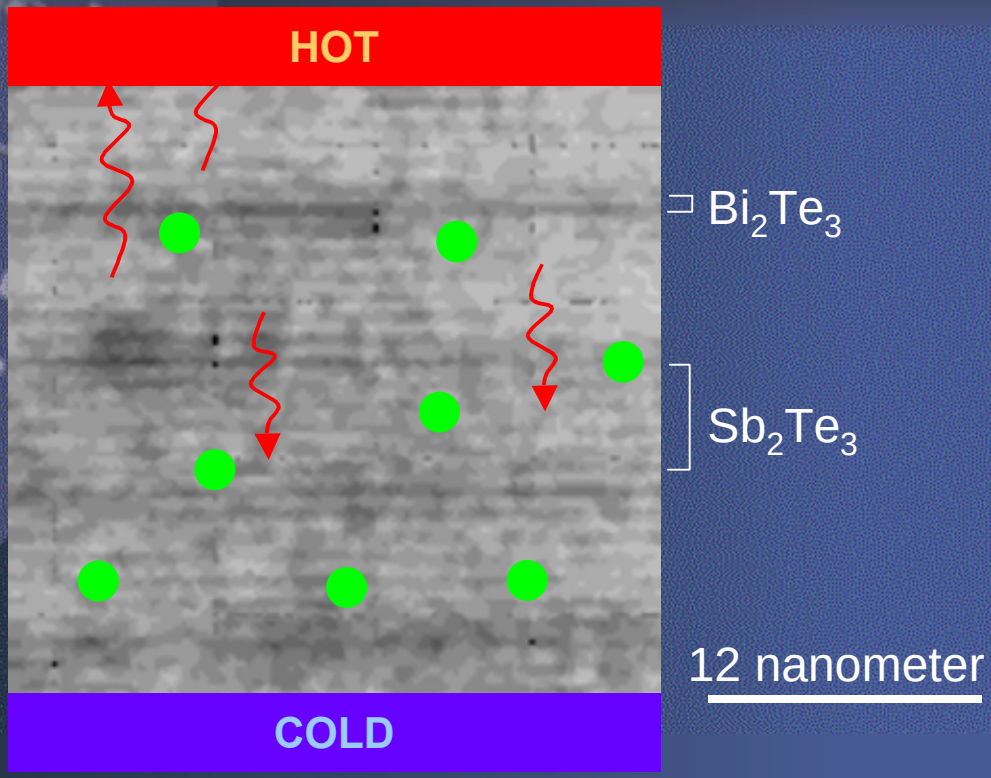

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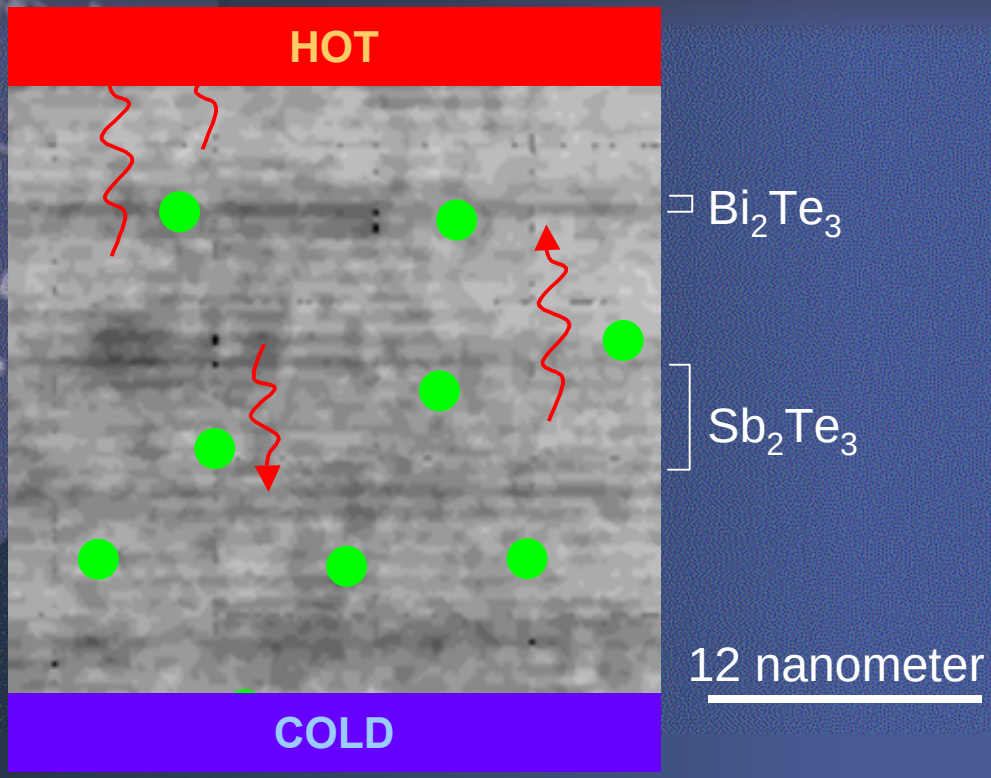

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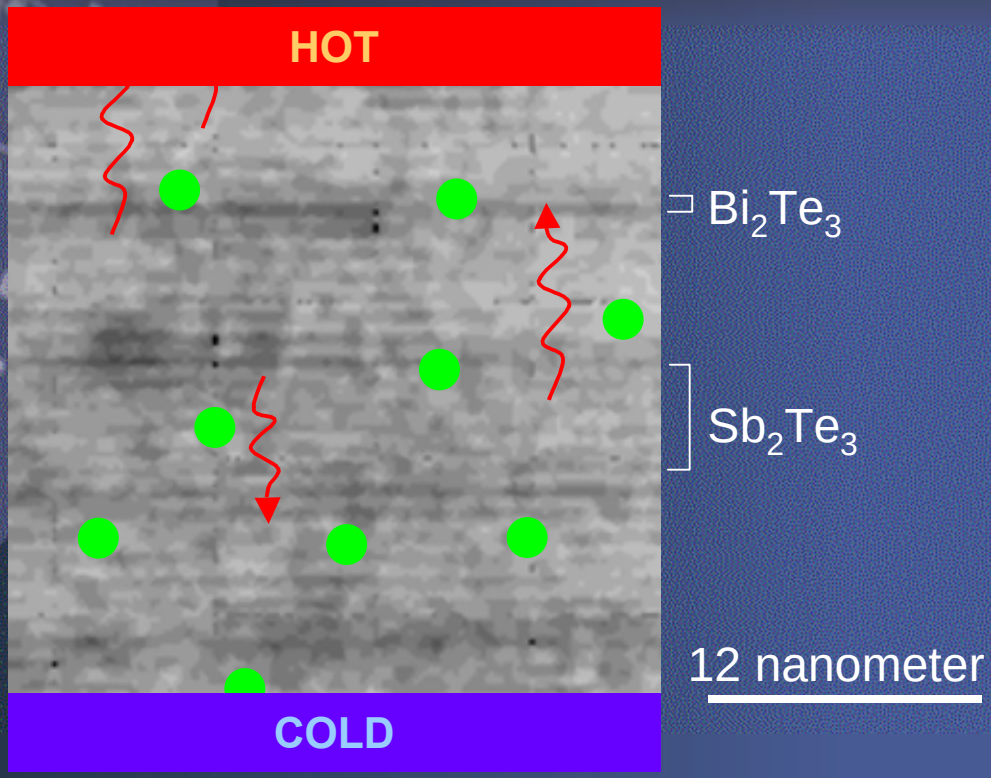

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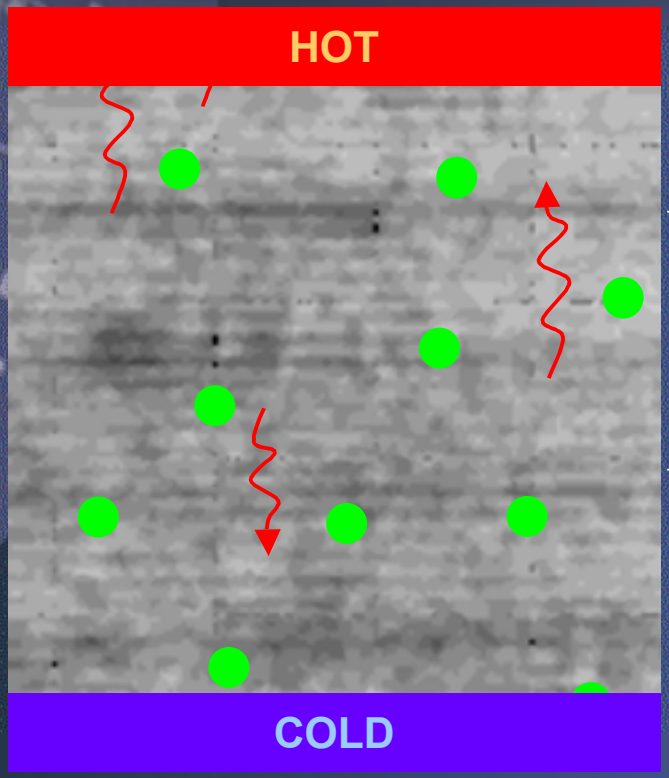

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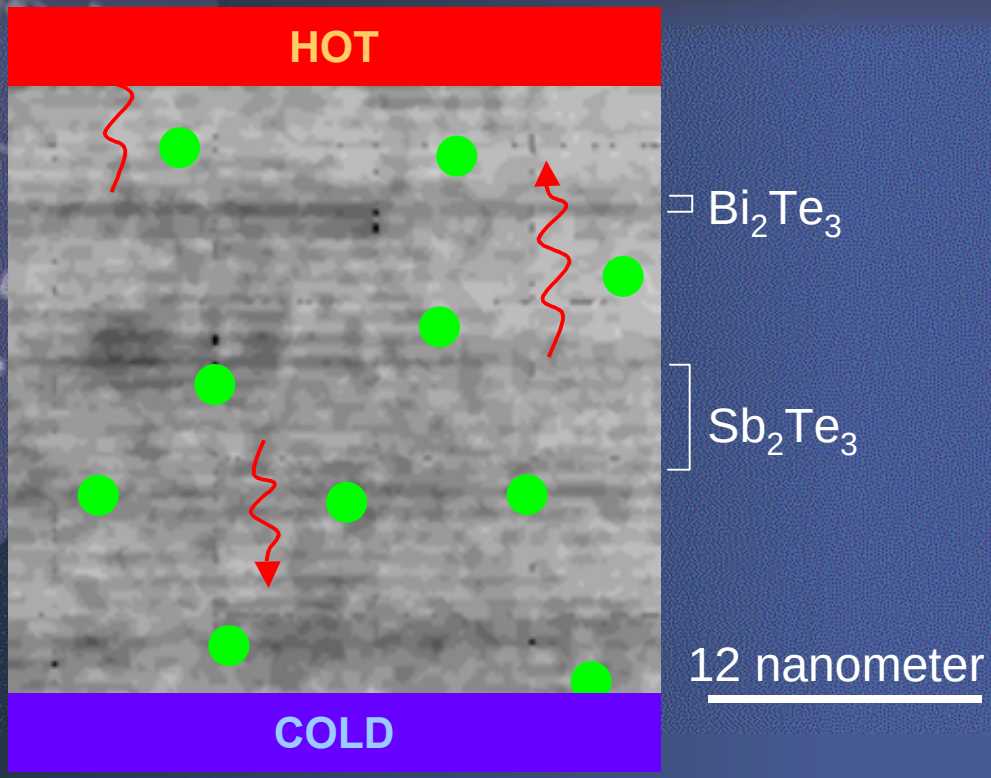

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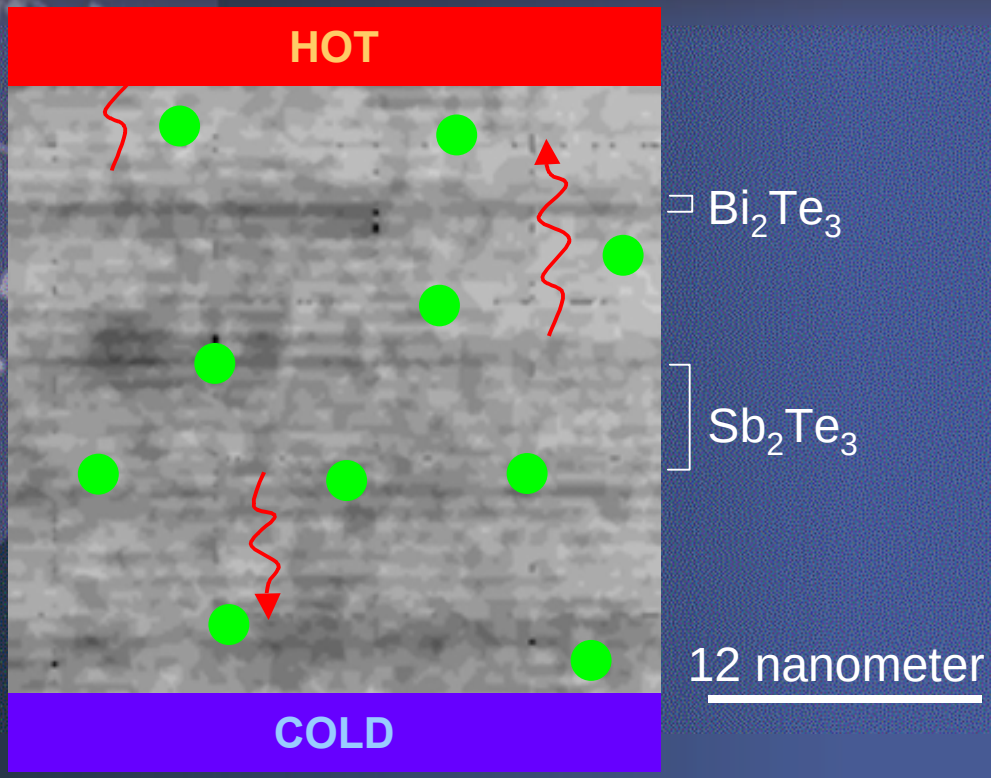

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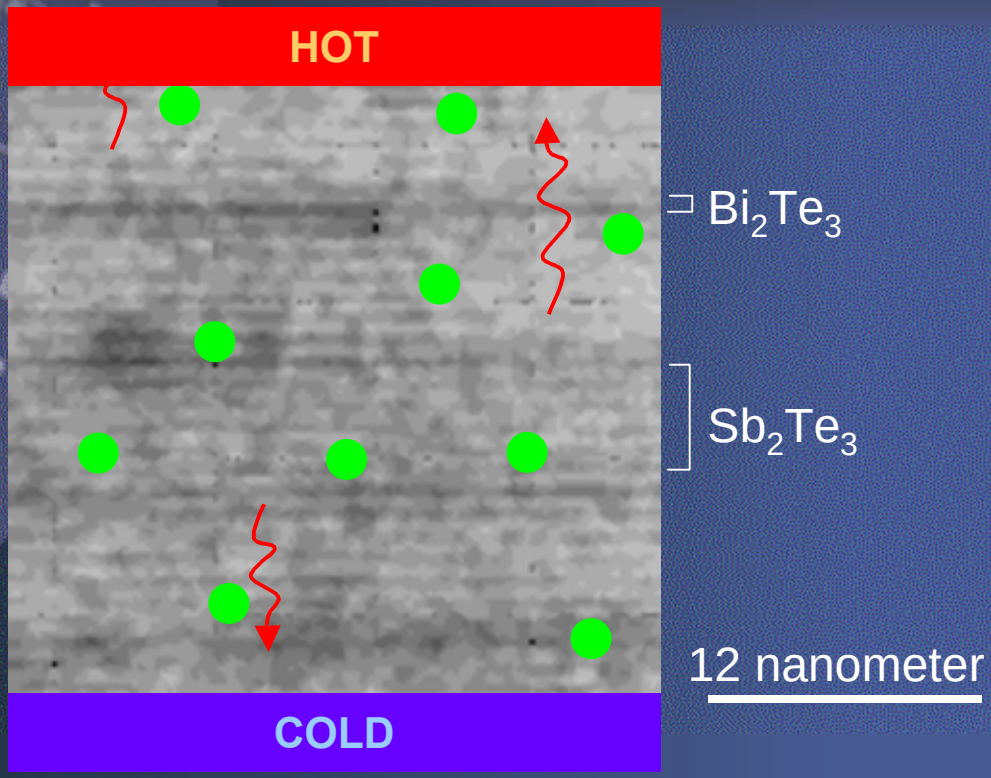

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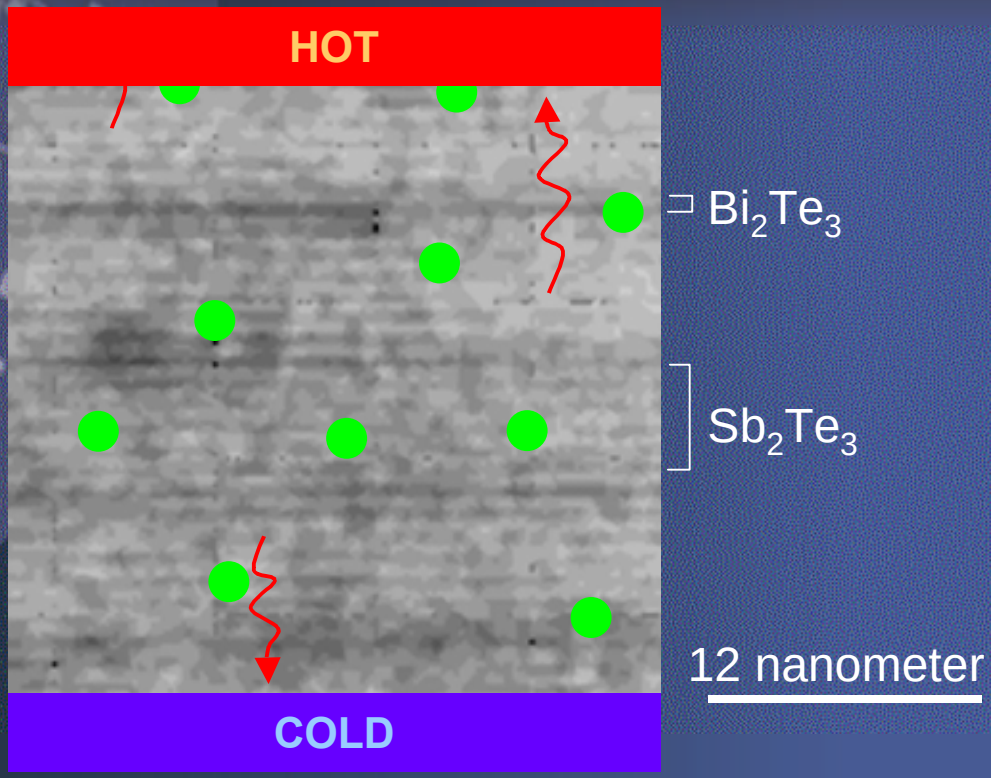

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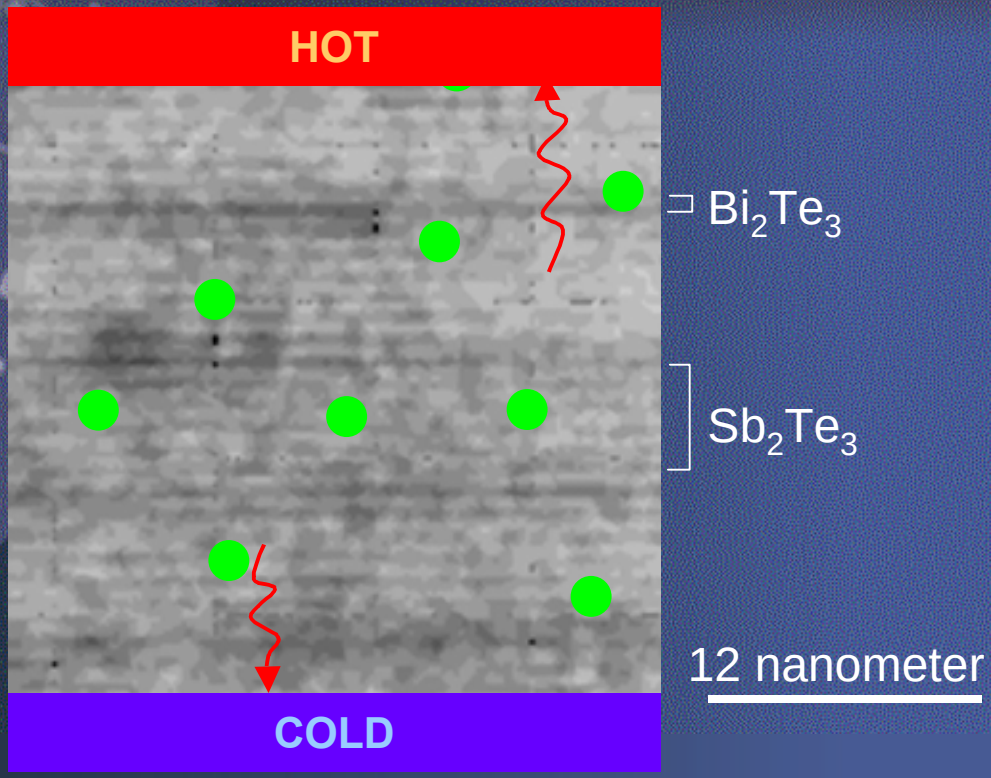

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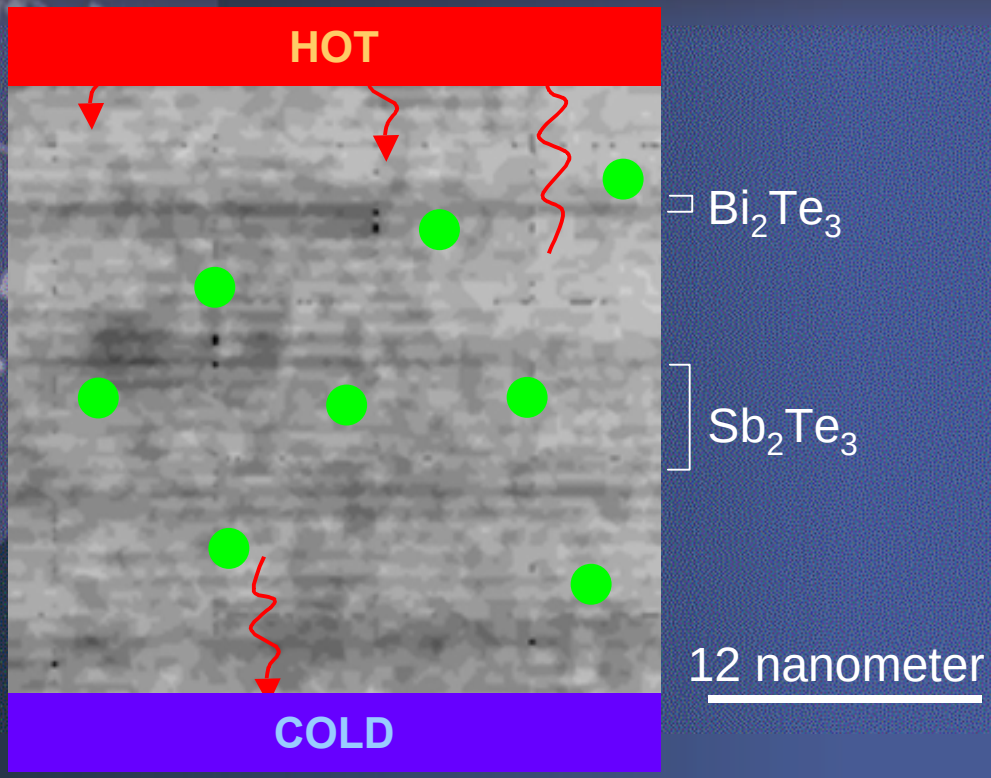

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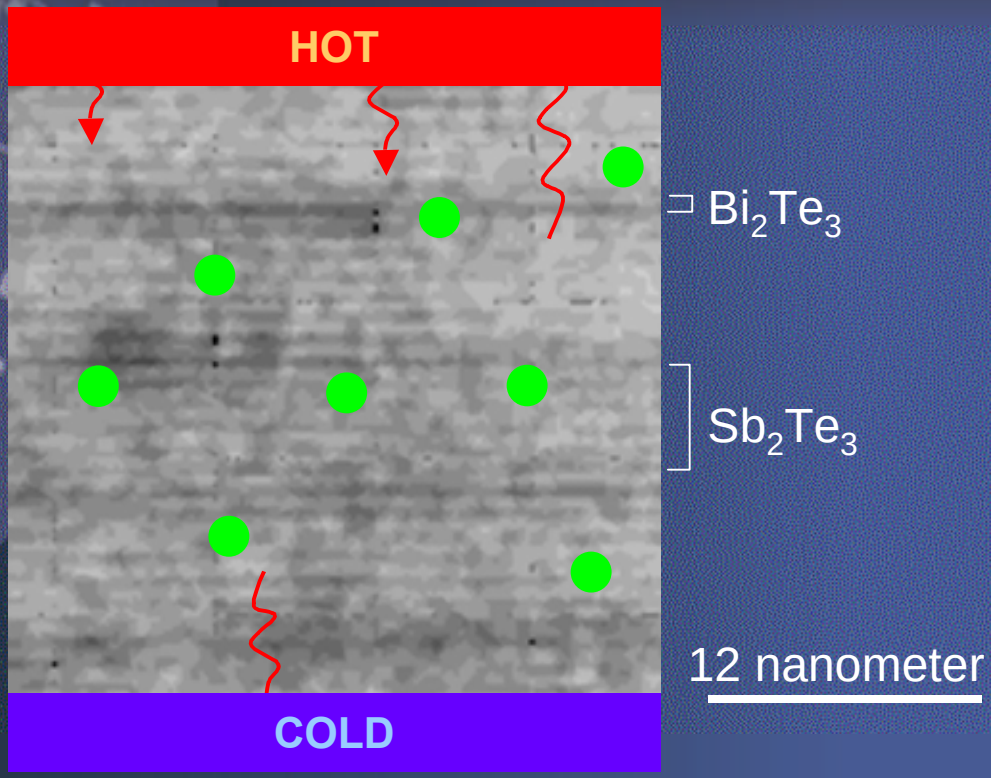

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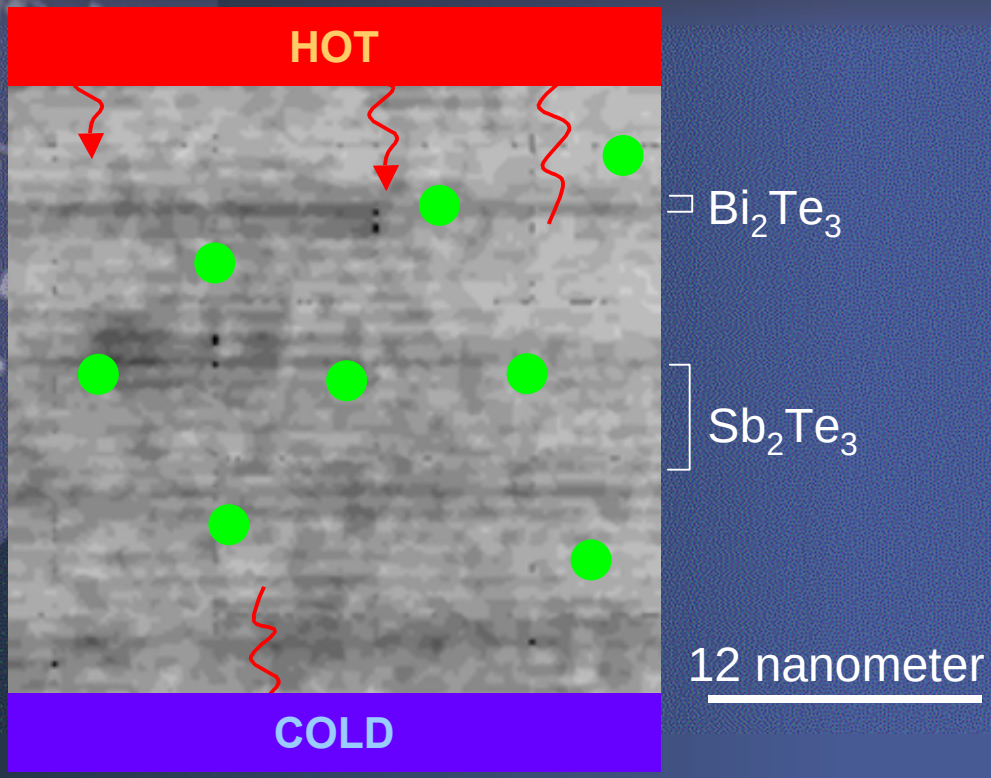

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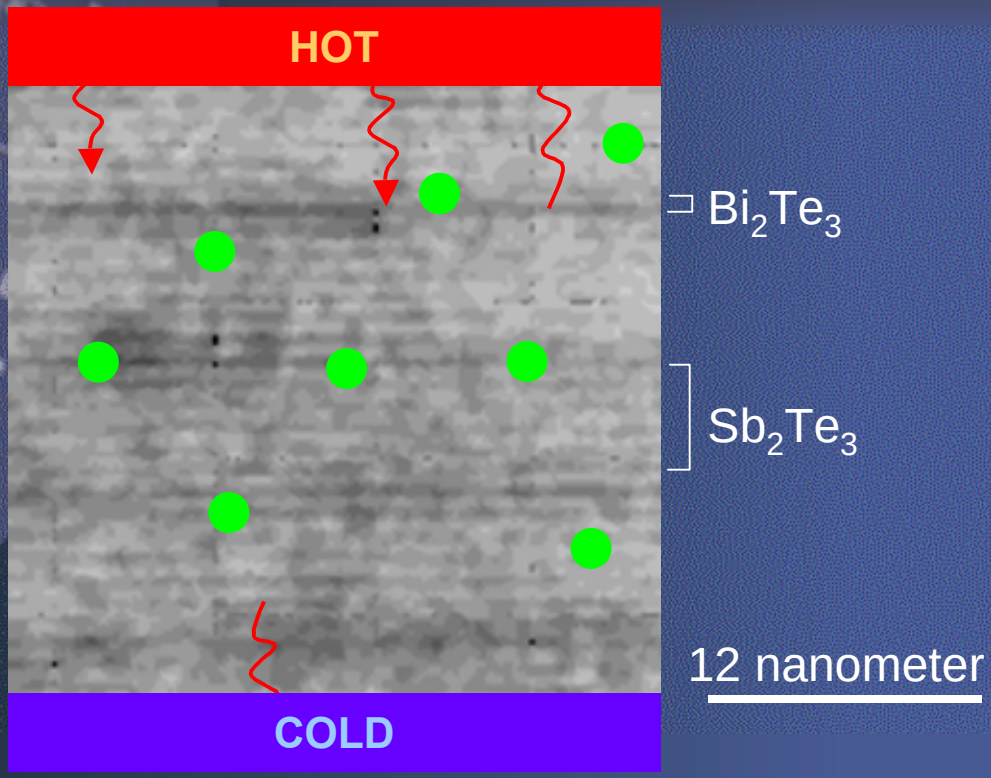

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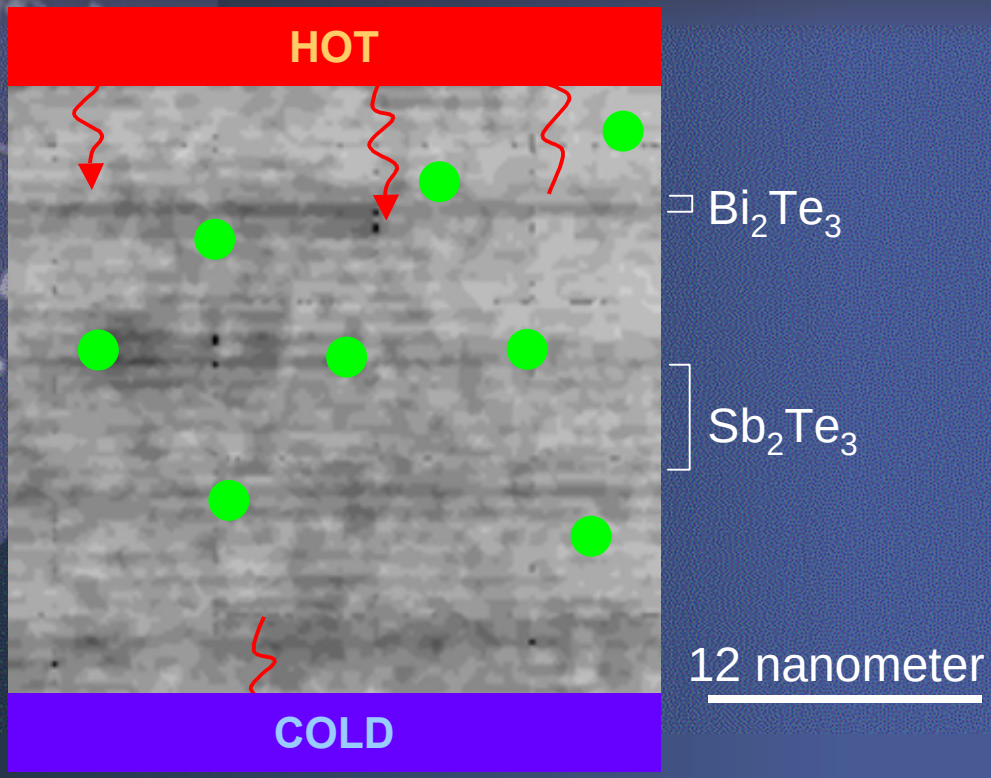

The RTI breakthrough arises from reducing heat transmission without disrupting electron flow

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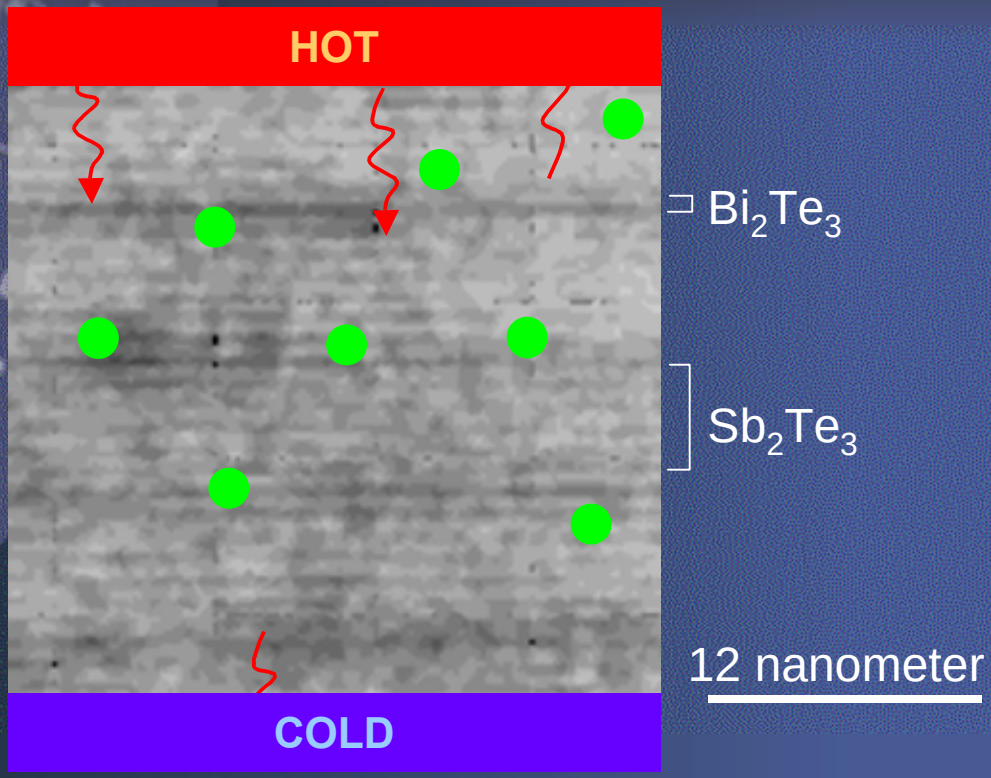

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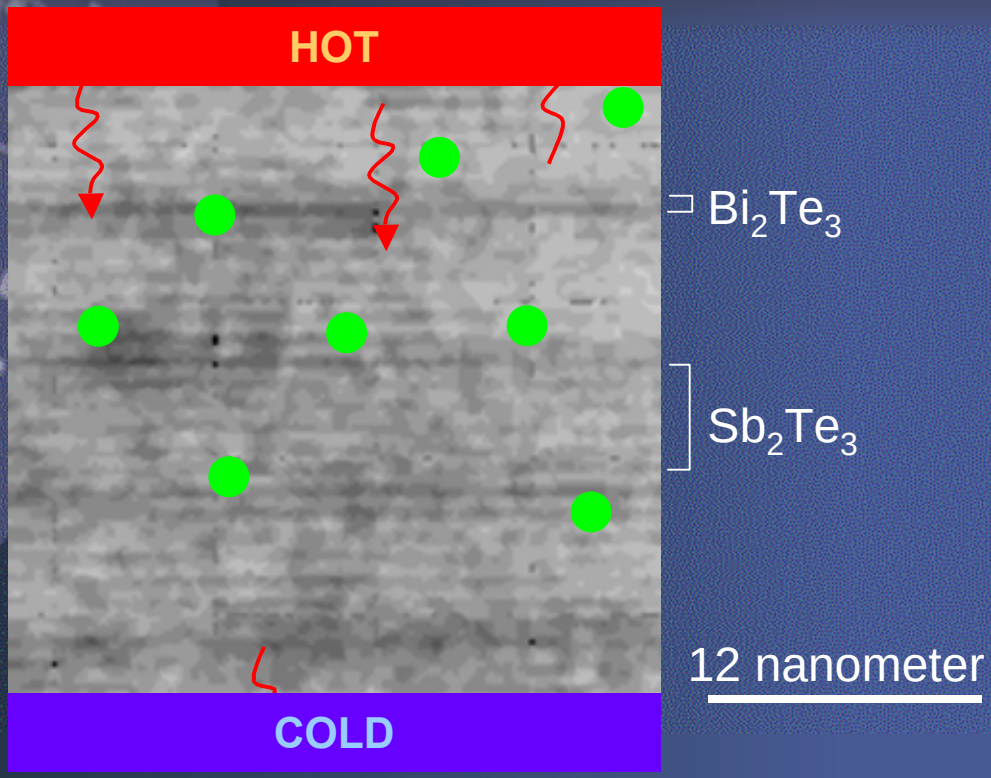

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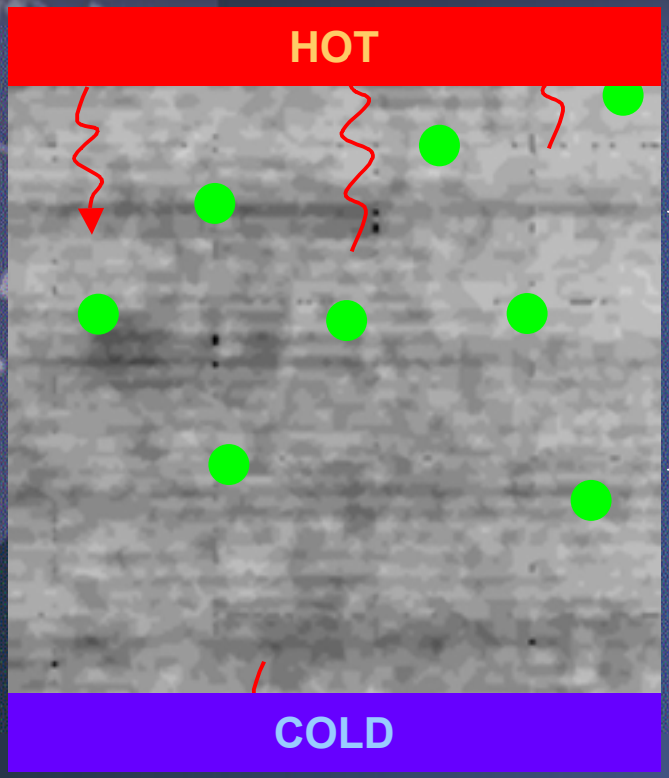


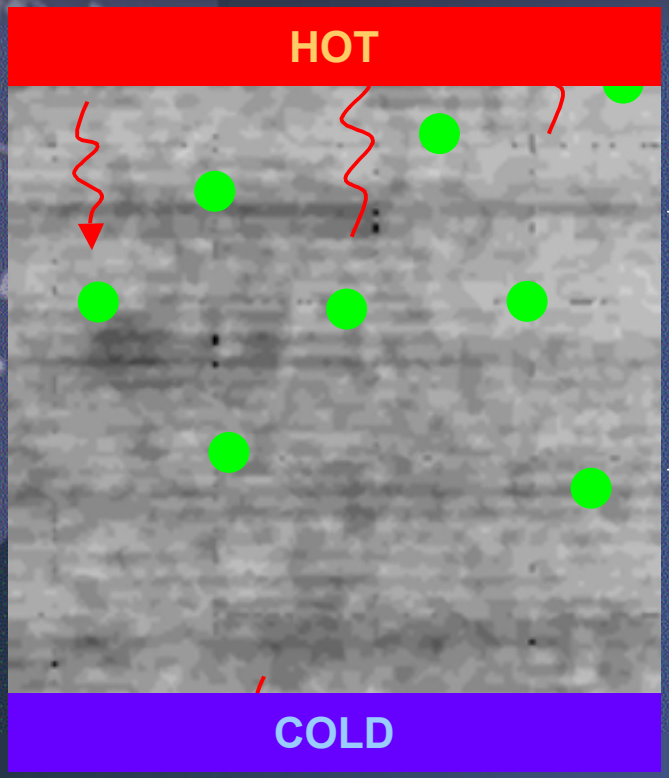
Bi_2Te_3

Sb_2Te_3

12 nanometer

The RTI breakthrough arises from reducing heat transmission without disrupting electron flow

RTI's Superlattice Material



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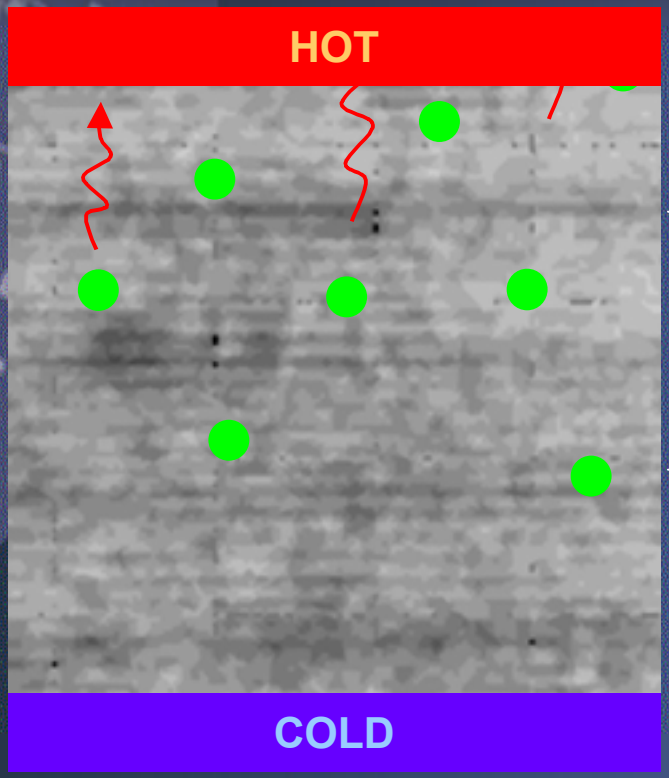

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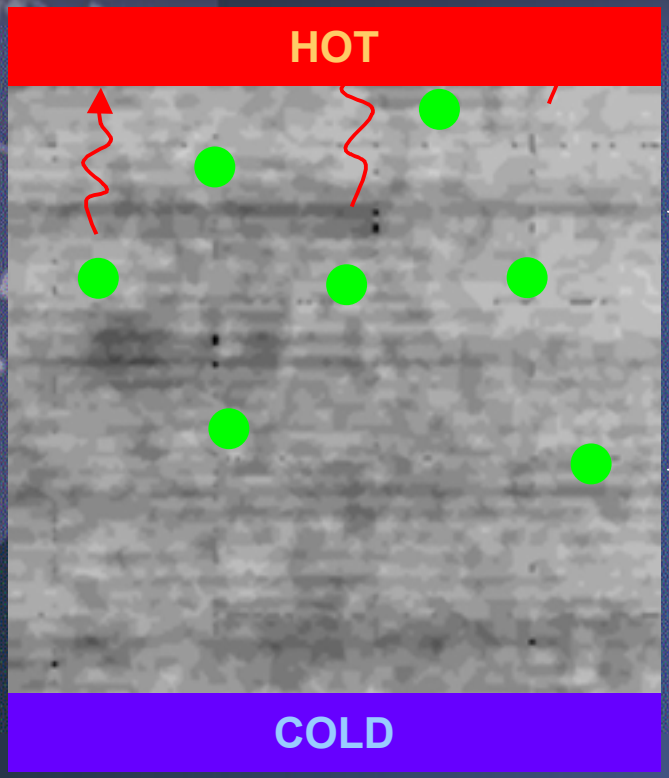
RTI's Superlattice Material



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12 nanometer

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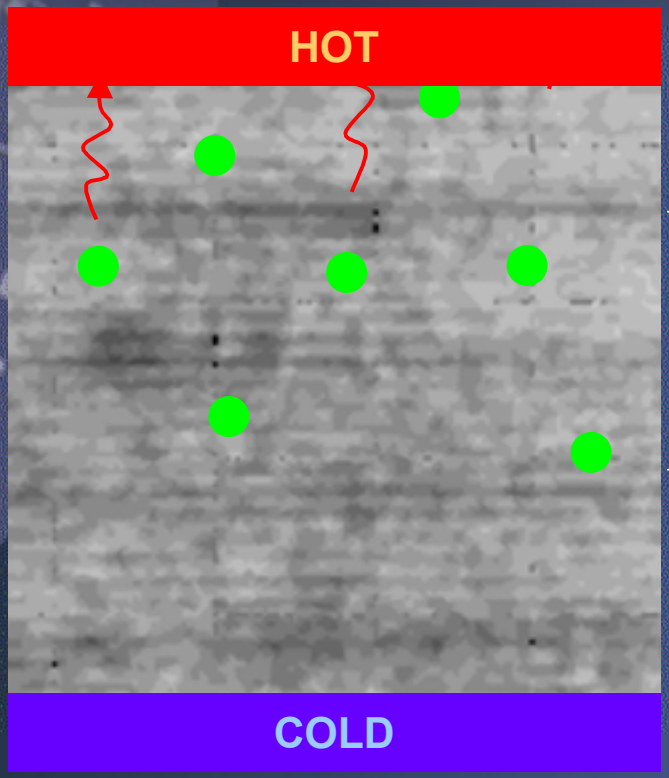

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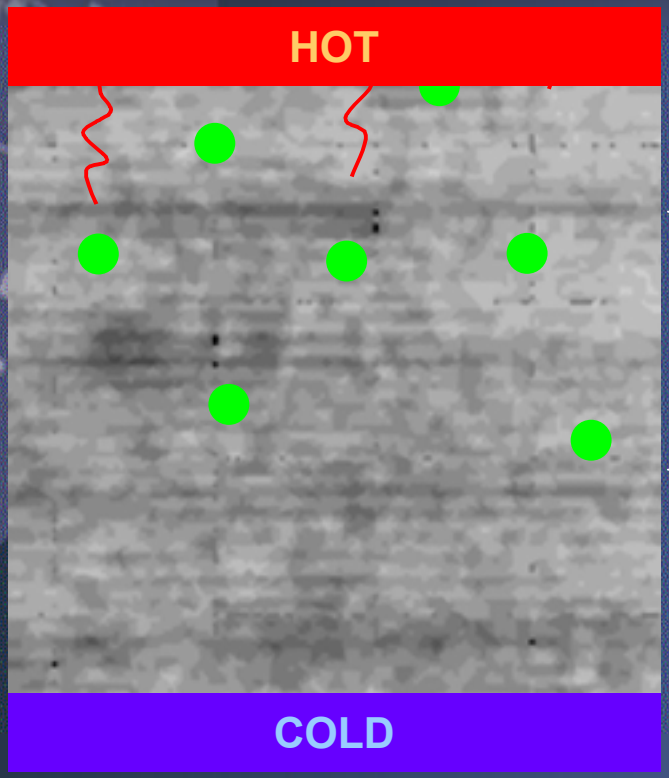

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Sb_2Te_3

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The RTI breakthrough arises from reducing heat transmission without disrupting electron flow

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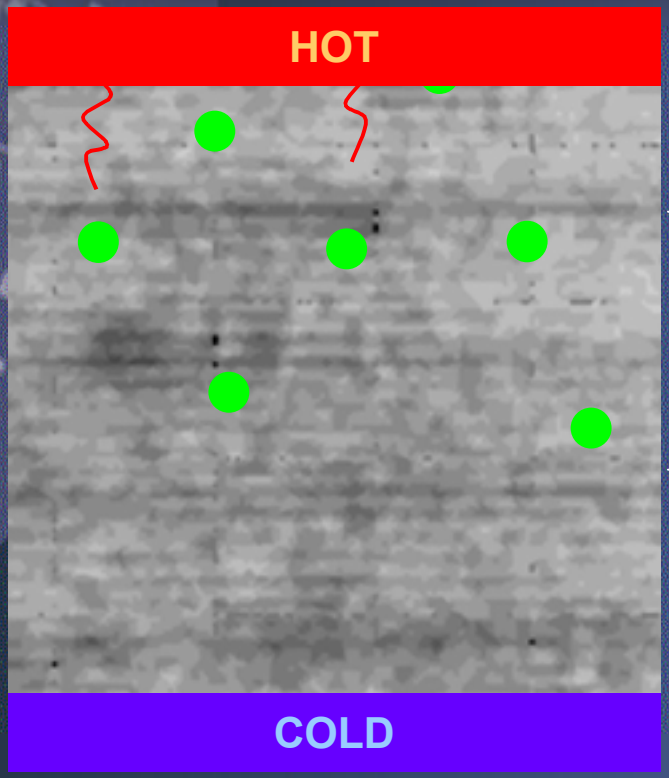

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Sb_2Te_3

12 nanometer

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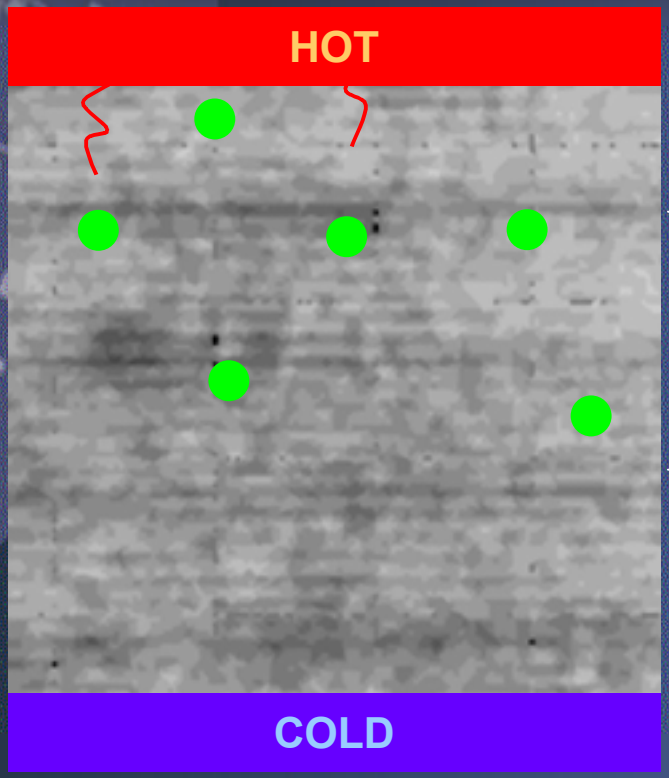
RTI's Superlattice Material



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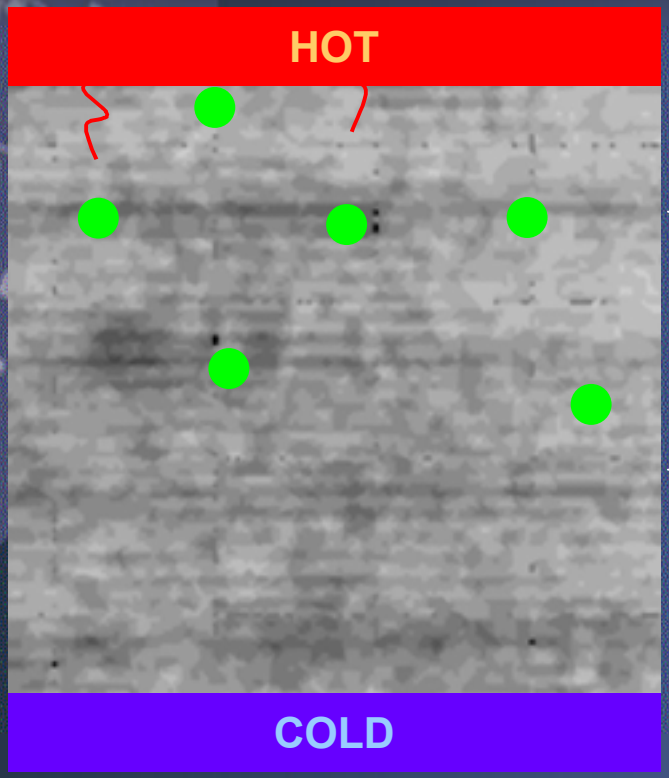
RTI's Superlattice Material



Bi_2Te_3

Sb_2Te_3

12 nanometer

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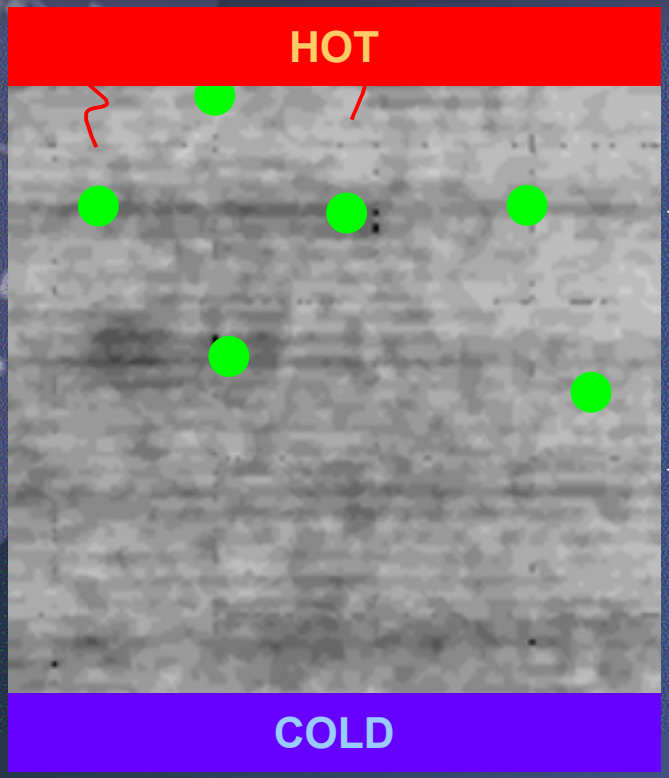
RTI's Superlattice Material



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Sb_2Te_3

12 nanometer

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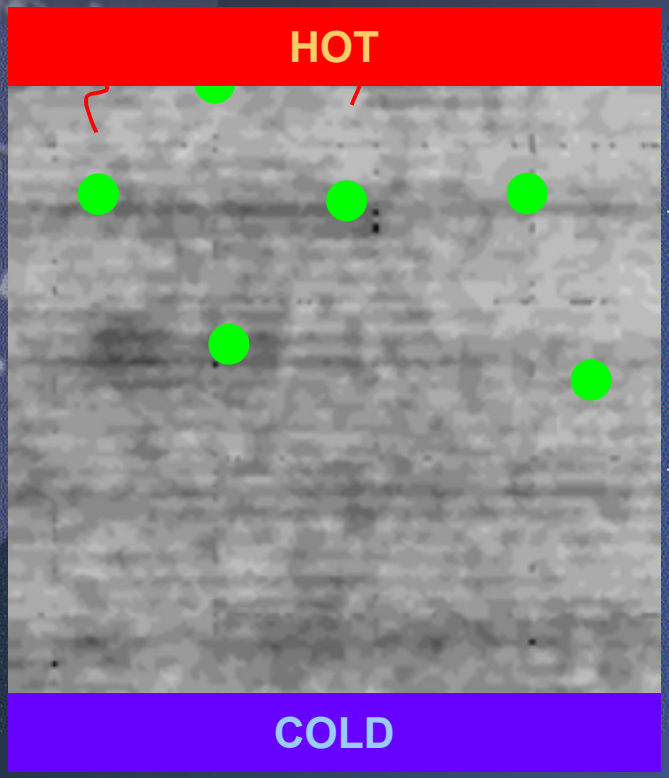
RTI's Superlattice Material



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Sb_2Te_3

12 nanometer

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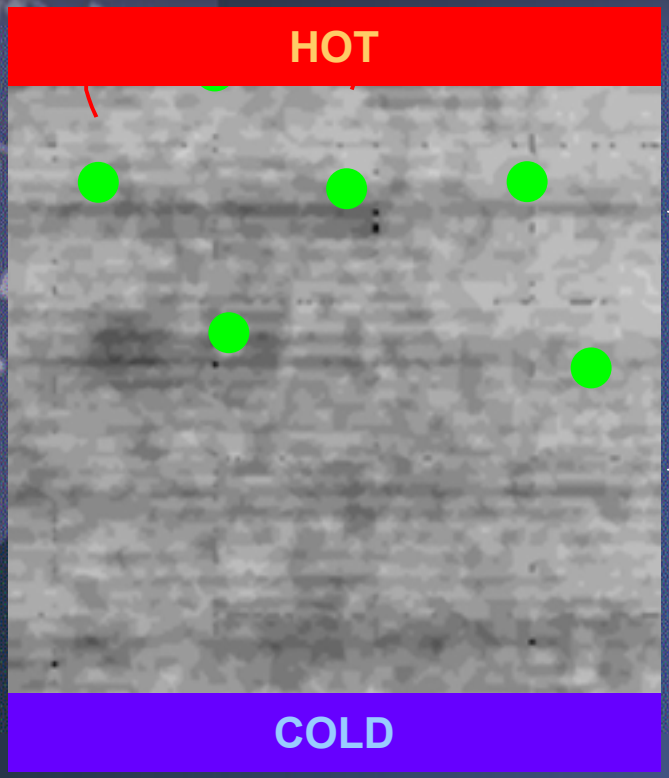

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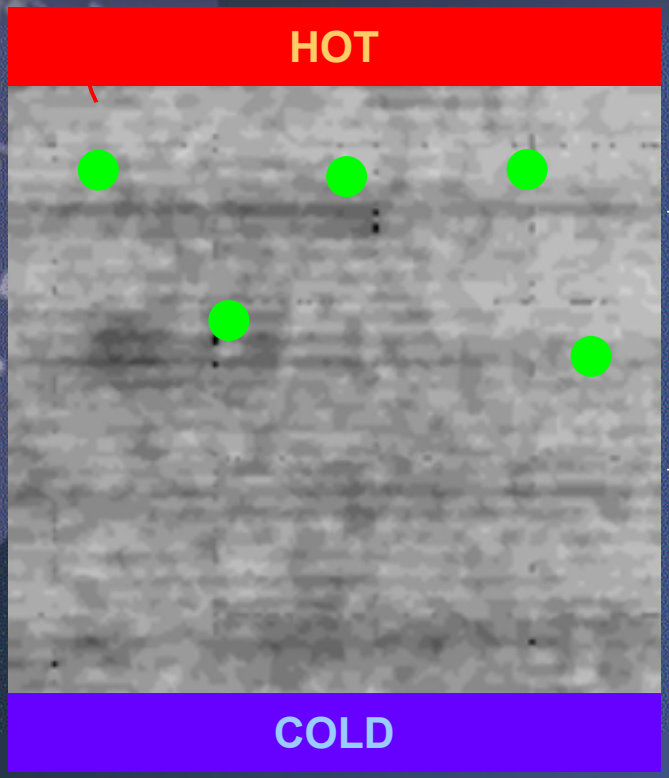

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Sb_2Te_3

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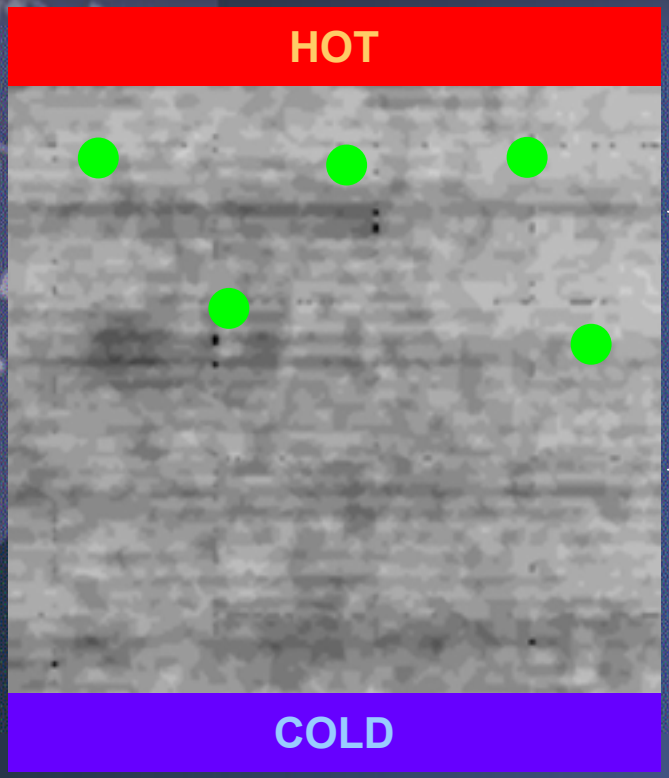


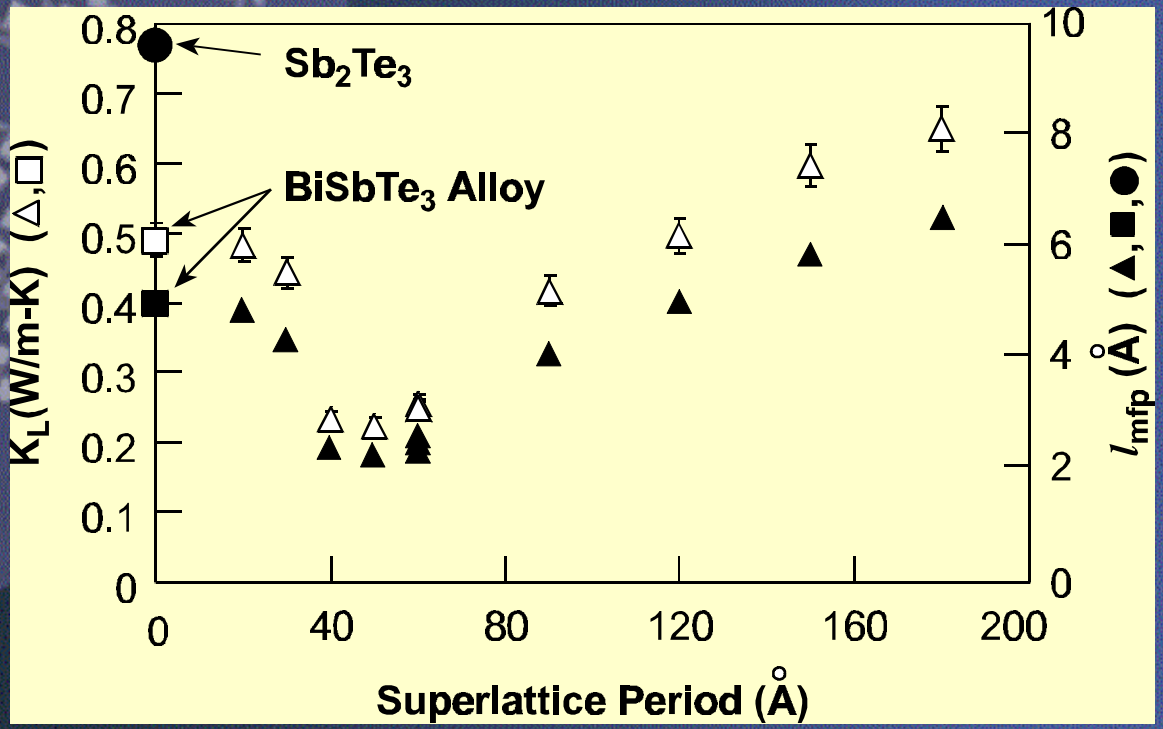
Bi_2Te_3

Sb_2Te_3

12 nanometer

The RTI breakthrough arises from reducing heat transmission without disrupting electron flow

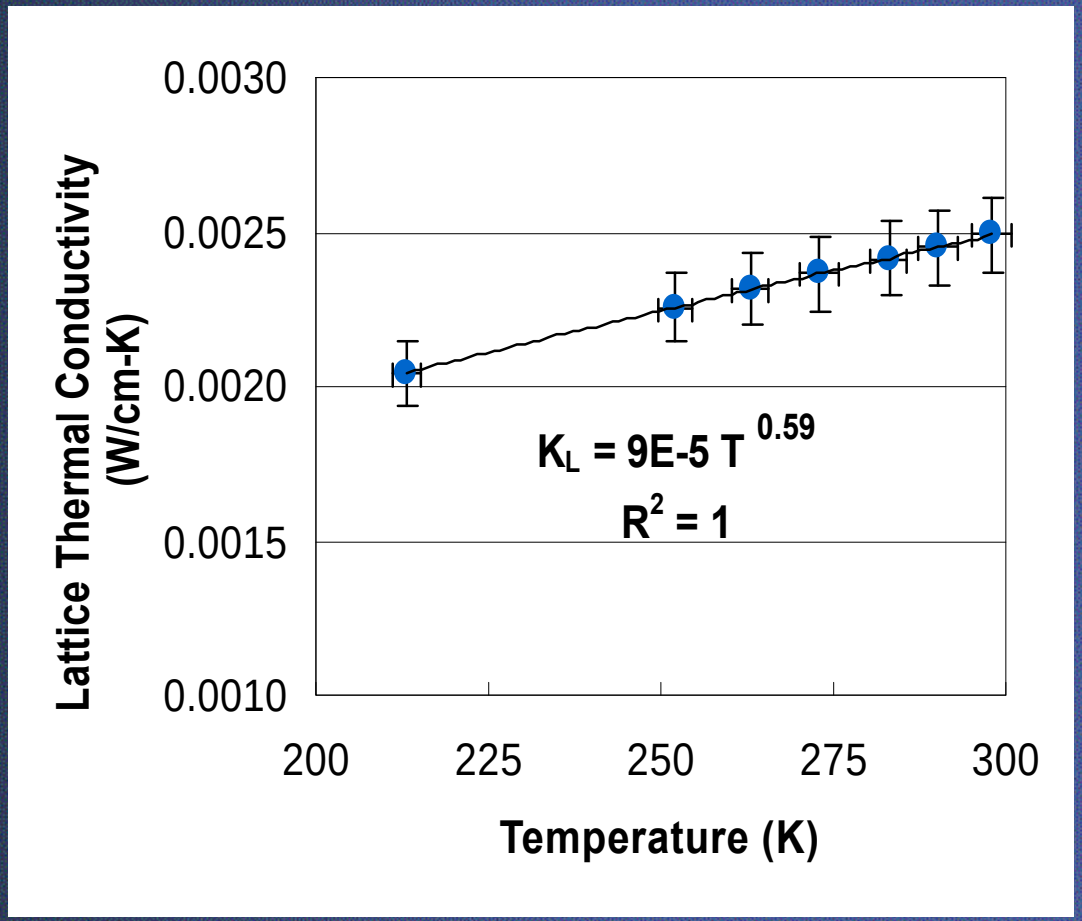
Thermal Conductivity Reduction in Bi_2Te_3/Sb_2Te_3 Superlattices



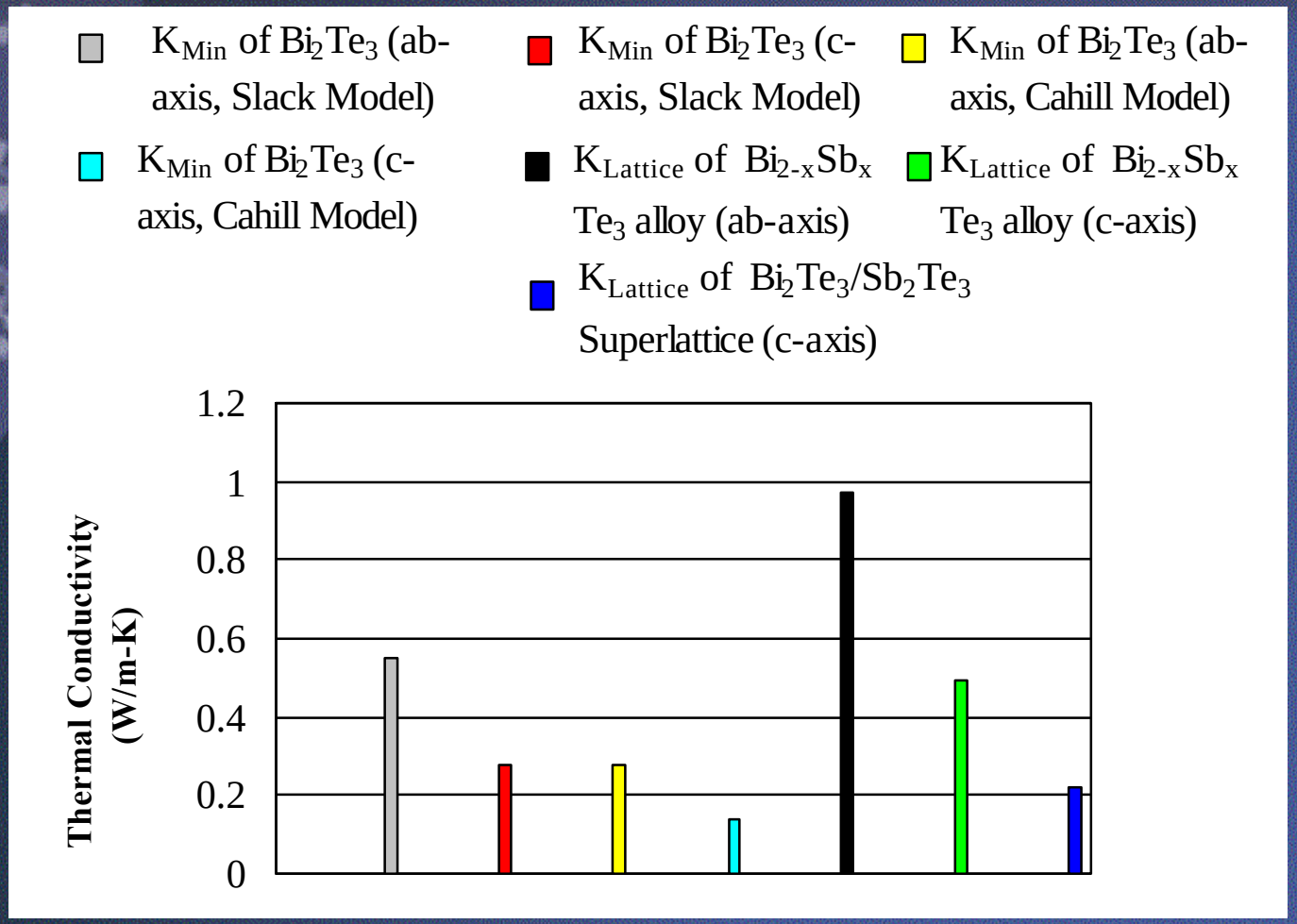
Physical Review B, 61, 3091 (2000)

- See Mahan et al., PRL, 84, 927, (2000)
  - Predicts a minimum although at a much smaller period

Temperature dependence of Lattice Thermal Conductivity in Bi_2Te_3/Sb_2Te_3 Superlattices

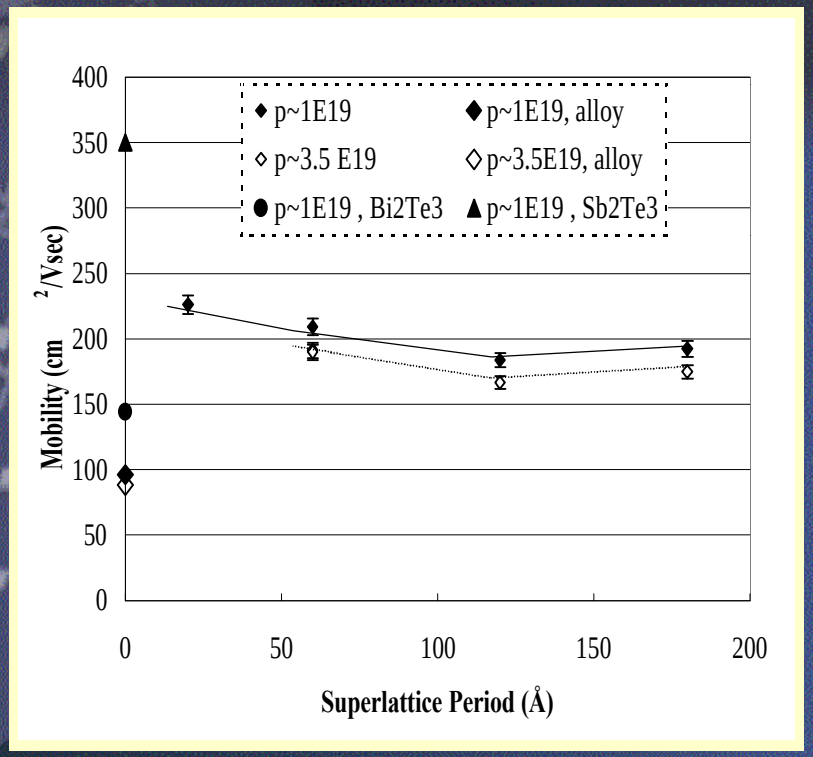


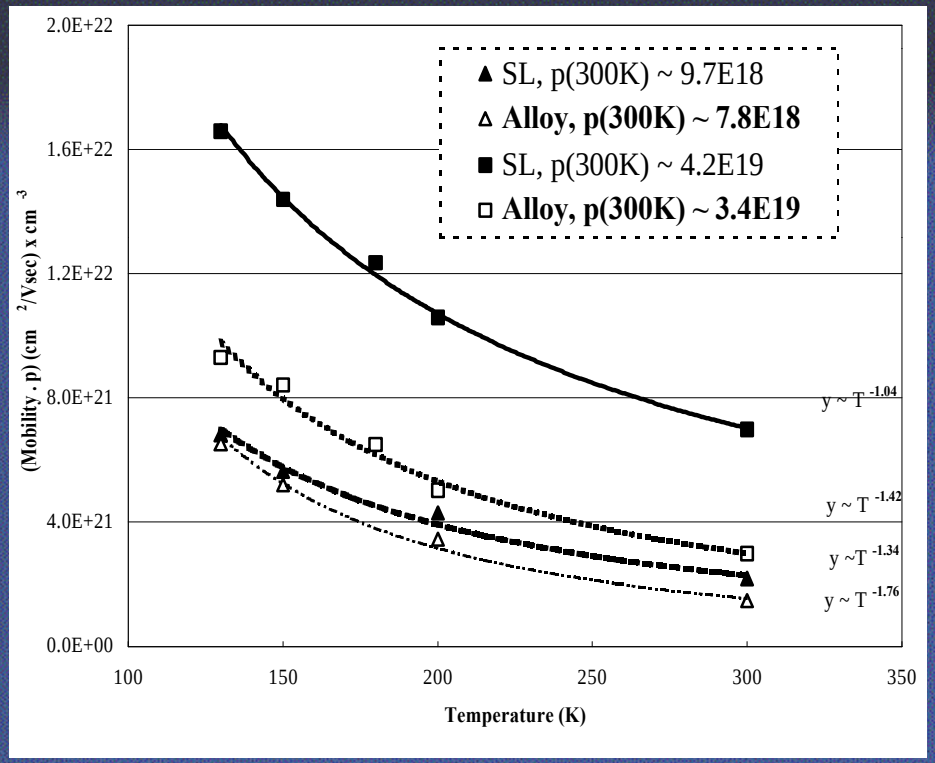
Lattice Thermal Conductivity in Bi_2Te_3/Sb_2Te_3 Superlattices compared to Minimum Thermal Conductivity in Bi_2Te_3



Nature, 413, 597 (2001)

In-Plane Hole Transport in p -type Bi_2Te_3/Sb_2Te_3 Superlattices

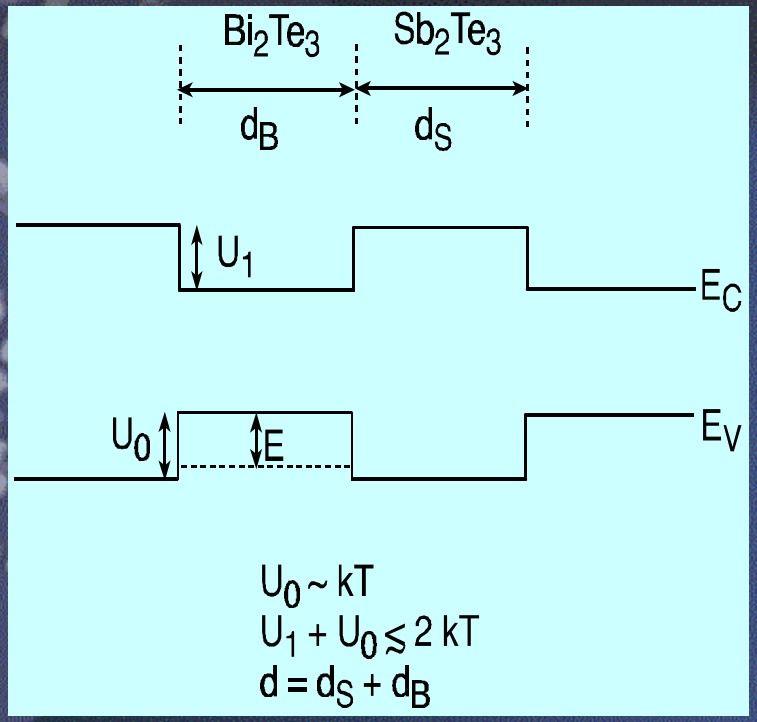


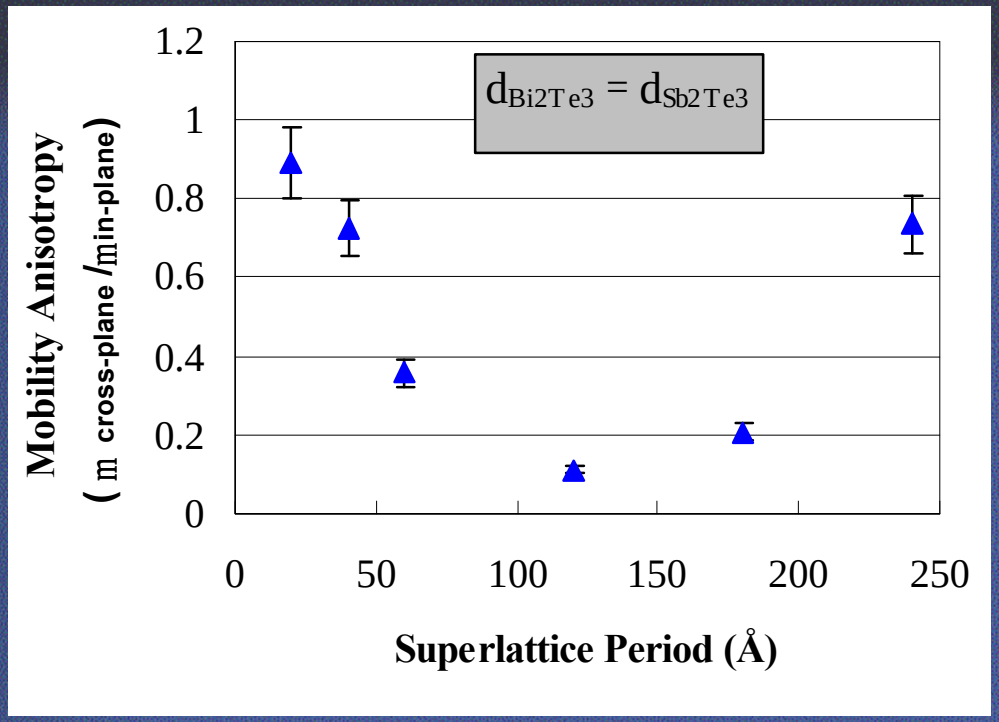


Appl. Phys. Lett., 75, 1104 (1999)

- **Enhanced In-Plane Mobilities in Superlattices from avoiding alloy scattering of carriers at 300K**
- **Indicates very little interface roughness from electronic (hole) scattering point-of-view**

Cross-Plane Hole Mini-band Transport in p -type Bi_2Te_3/Sb_2Te_3 Superlattices

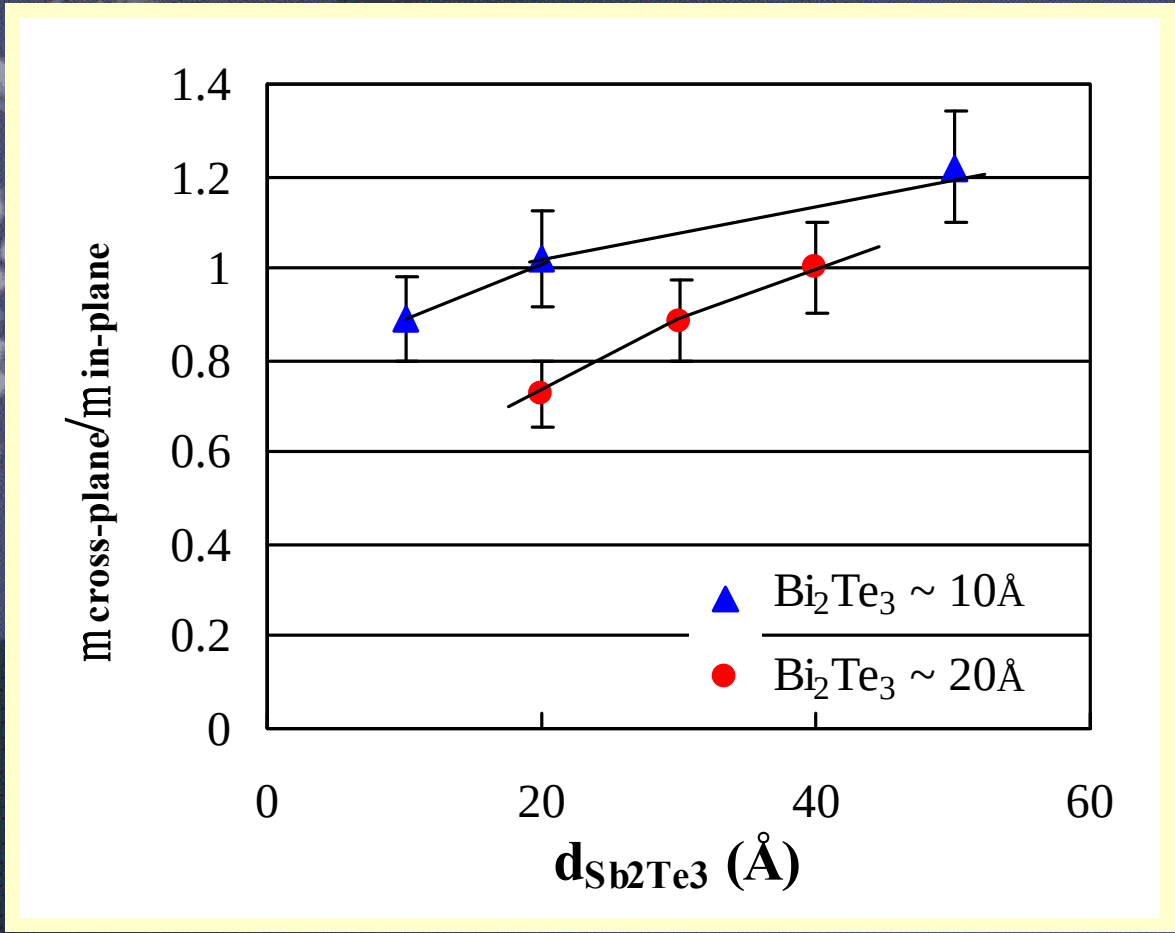




Nature, 413, 597 (2001)

- $\Delta E_v - E = (m^* d_B^2 \Delta E_v^{\ 2}) / 2\hbar^2$
- $2\Delta \sim \exp. \{-[(8m^*_{hh}/\hbar^2)(\Delta E_v - E)d_S]^{1/2}\}$
- $\mu_{cross\text{-}plane} \sim [e\,\Delta\, d^2/\hbar^2]\ \tau$
- Ref: Capasso et al., IEEE J. of Quantum Electronics, (1986)

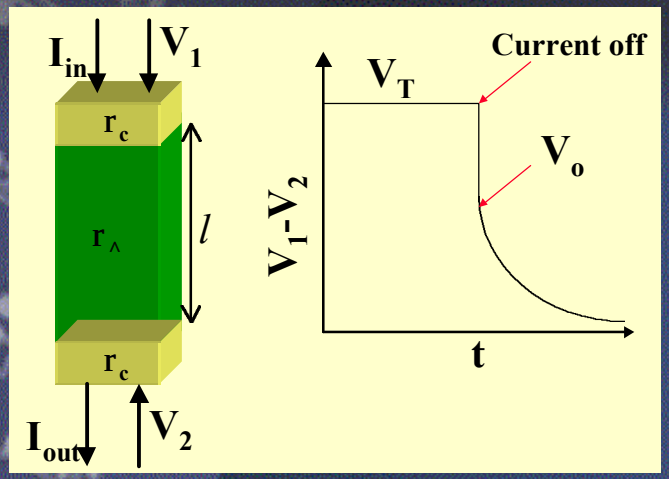
Cross-Plane Hole Transport in p -type Bi_2Te_3/Sb_2Te_3 Superlattices

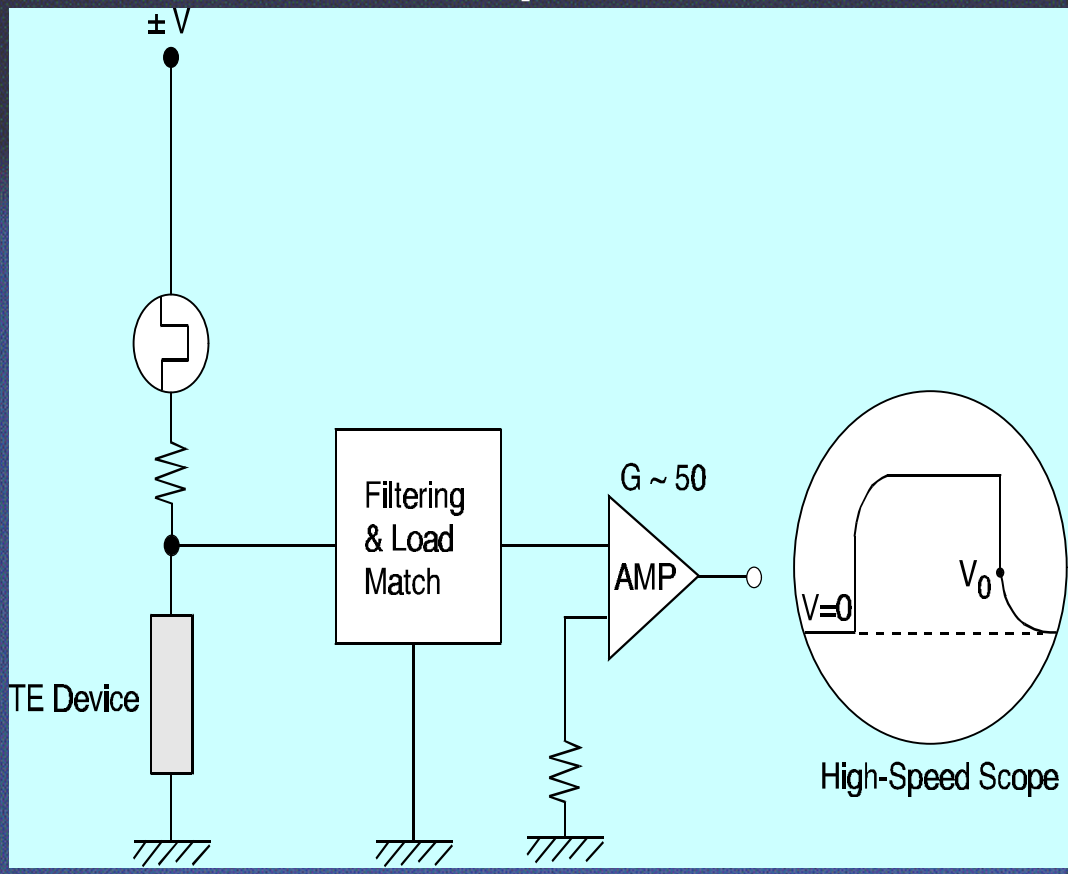


- **Enhanced Cross-Plane Mobilities as well in the superlattices**
- **Moderate to negligible electrical anisotropy**
  - **<u>From negligible quantum-confinement and weak mini-band conduction</u>**

Nature, 413, 597 (2001)

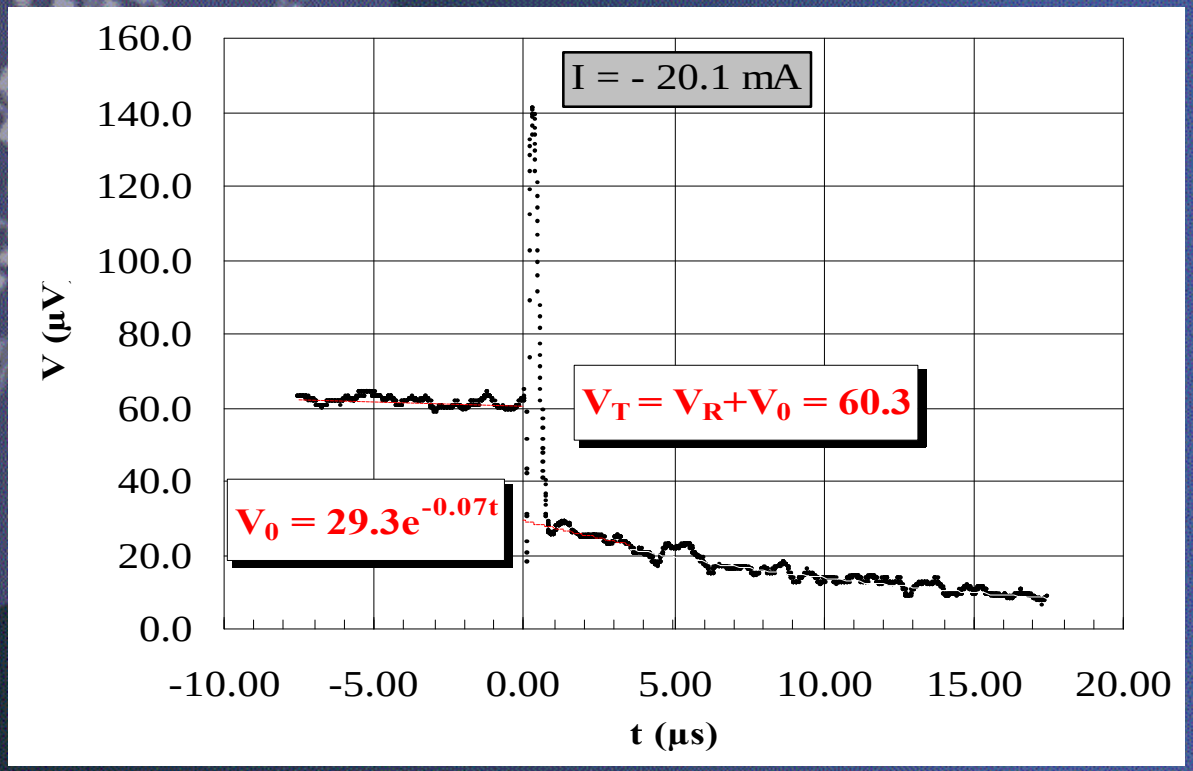
ZT Measurement Technique





- $ZT_{device} = V_o/V_T - V_o$
  $= V_o/V_R$
  = Peltier Voltage/Ohmic Voltage
- All properties that make up ZT are measured at the same place, same time with current flowing
- Difficult as it involves minimizing contact resistances
- $ZT_{device} \sim ZT_{intrinsic} \{1+2R_{contacts}/R_{device}\}^{-1}$

ZT in p -type bulk material thinned down to 5 microns – same as that of our superlattice film

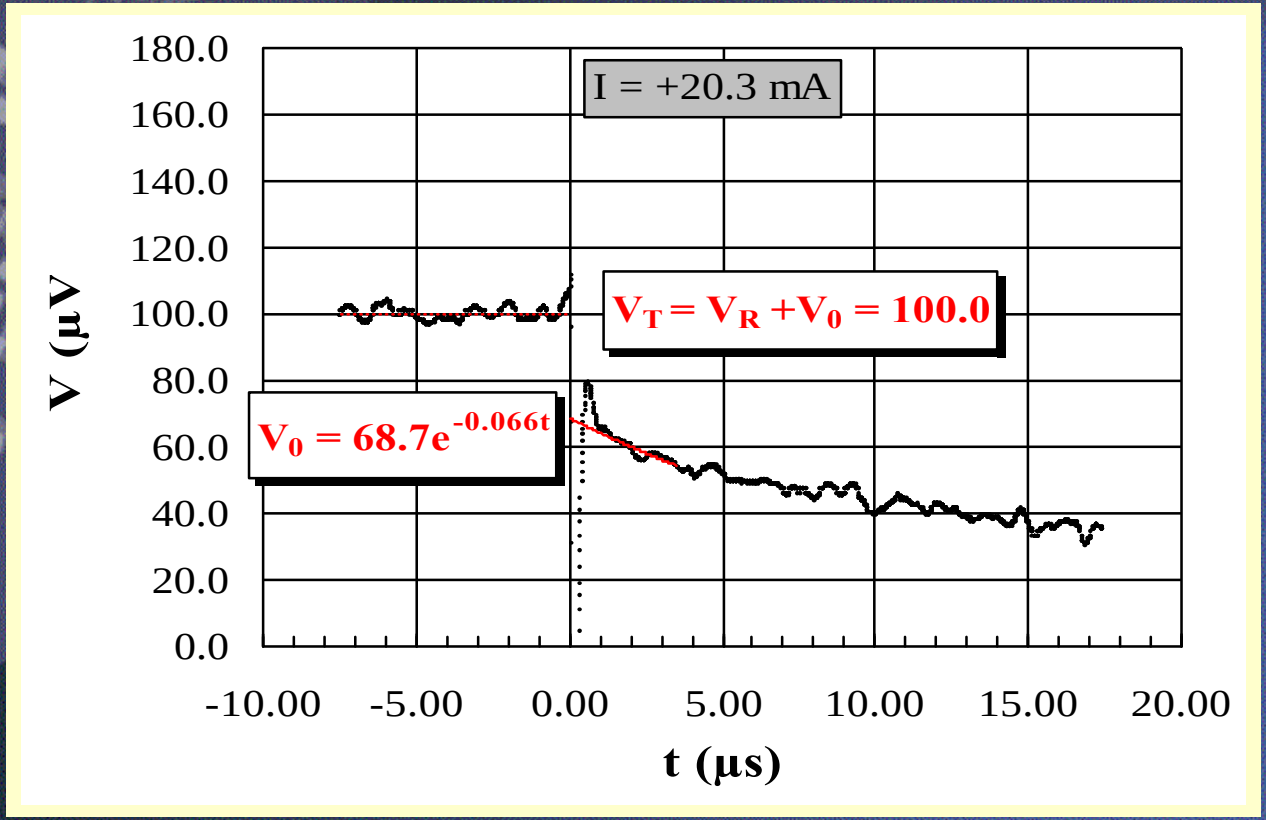


- ZT_{device}

  = 29.3/(60.3-29.3)

  = 0.94
- **From both current directions, ZT ~ 0.97 ± 0.03**
- **<u>No unusual effects because of the thin-film nature per se!</u>**

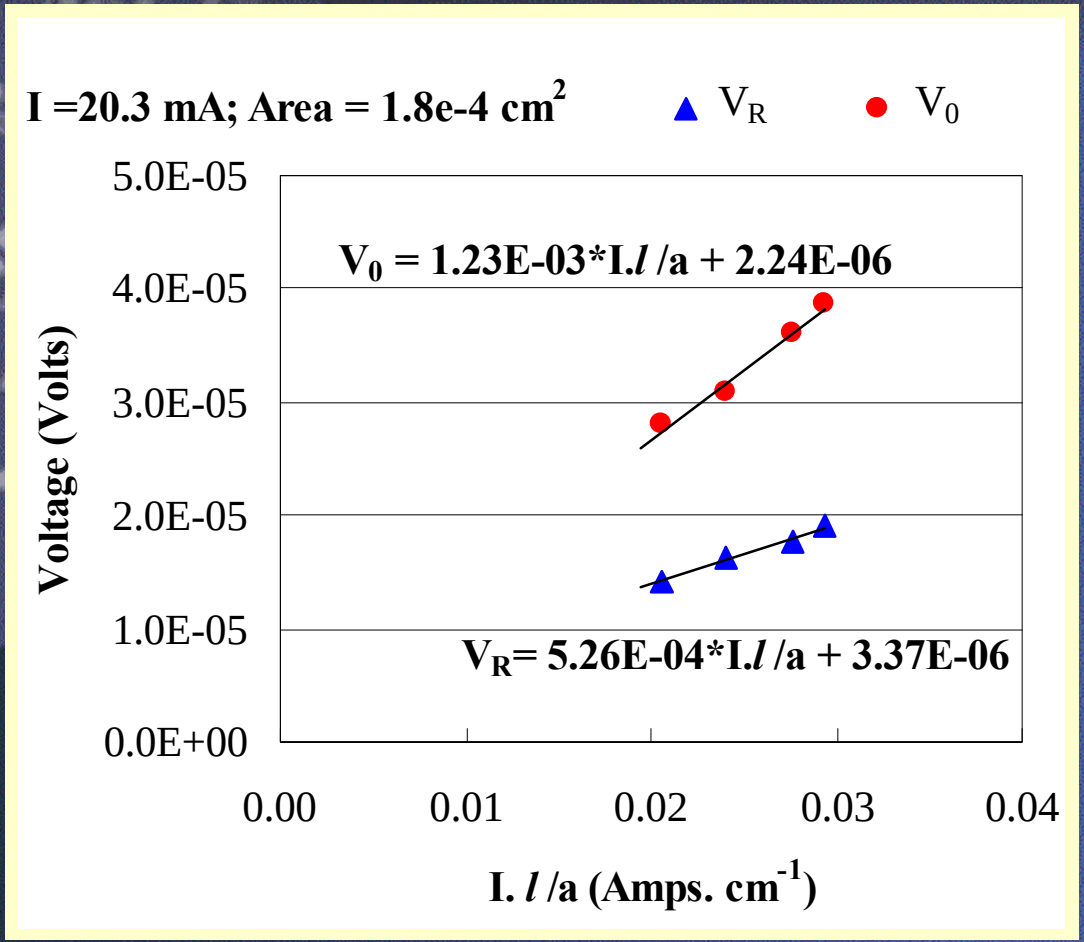
ZT in p -type Bi_2Te_3/Sb_2Te_3 Superlattices



- ZT_{device}
  = 68.7/(100.0-68.7)
  = 2.19
- **From both current directions, ZT ~2.39 ± 0.19**

Nature, 413, 597 (2001)

Intrinsic ZT in p -type Bi_2Te_3/Sb_2Te_3 Superlattices by Variable-Thickness Method

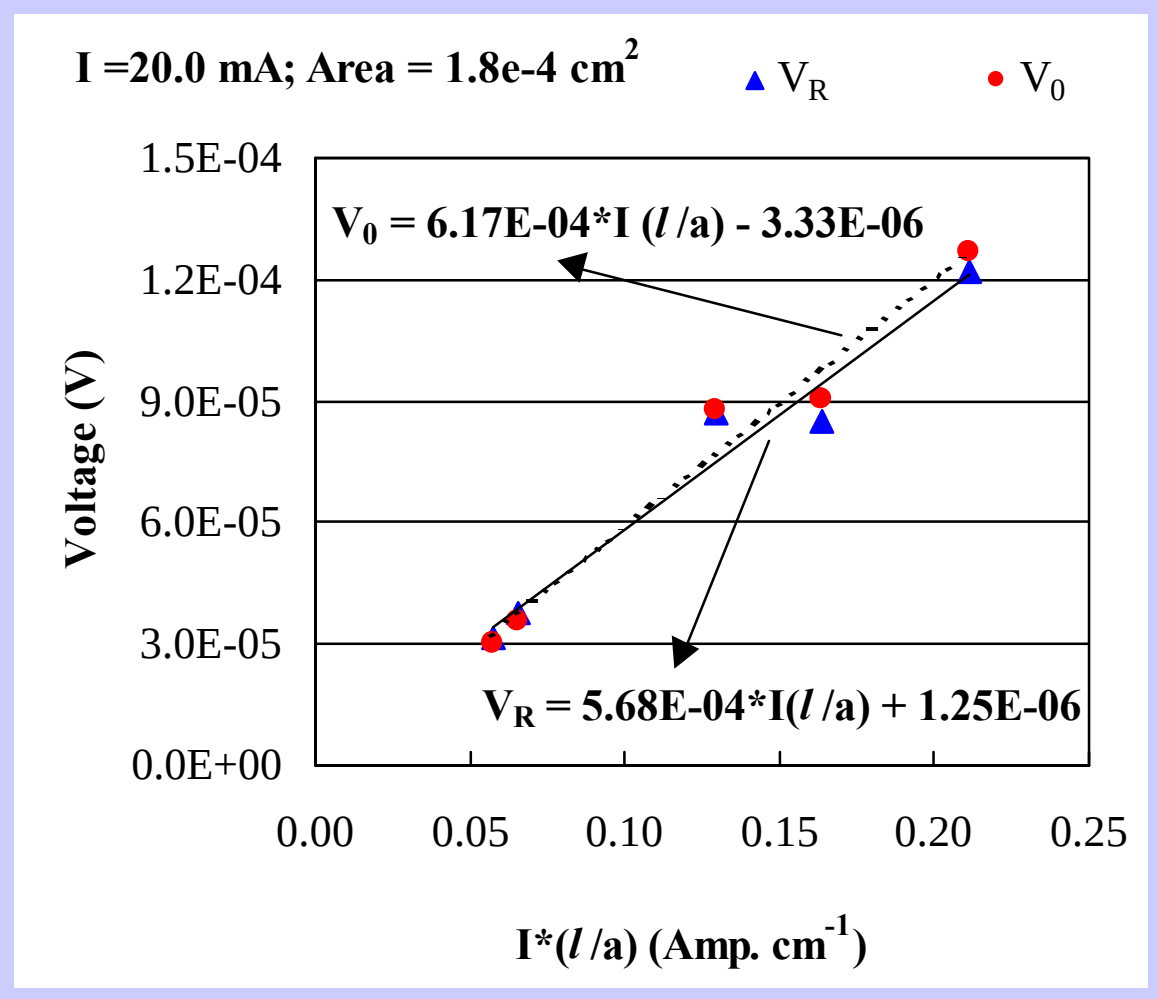


$ZT_{intrinsic} \sim 2.34$

Nature, 413, 597 (2001)

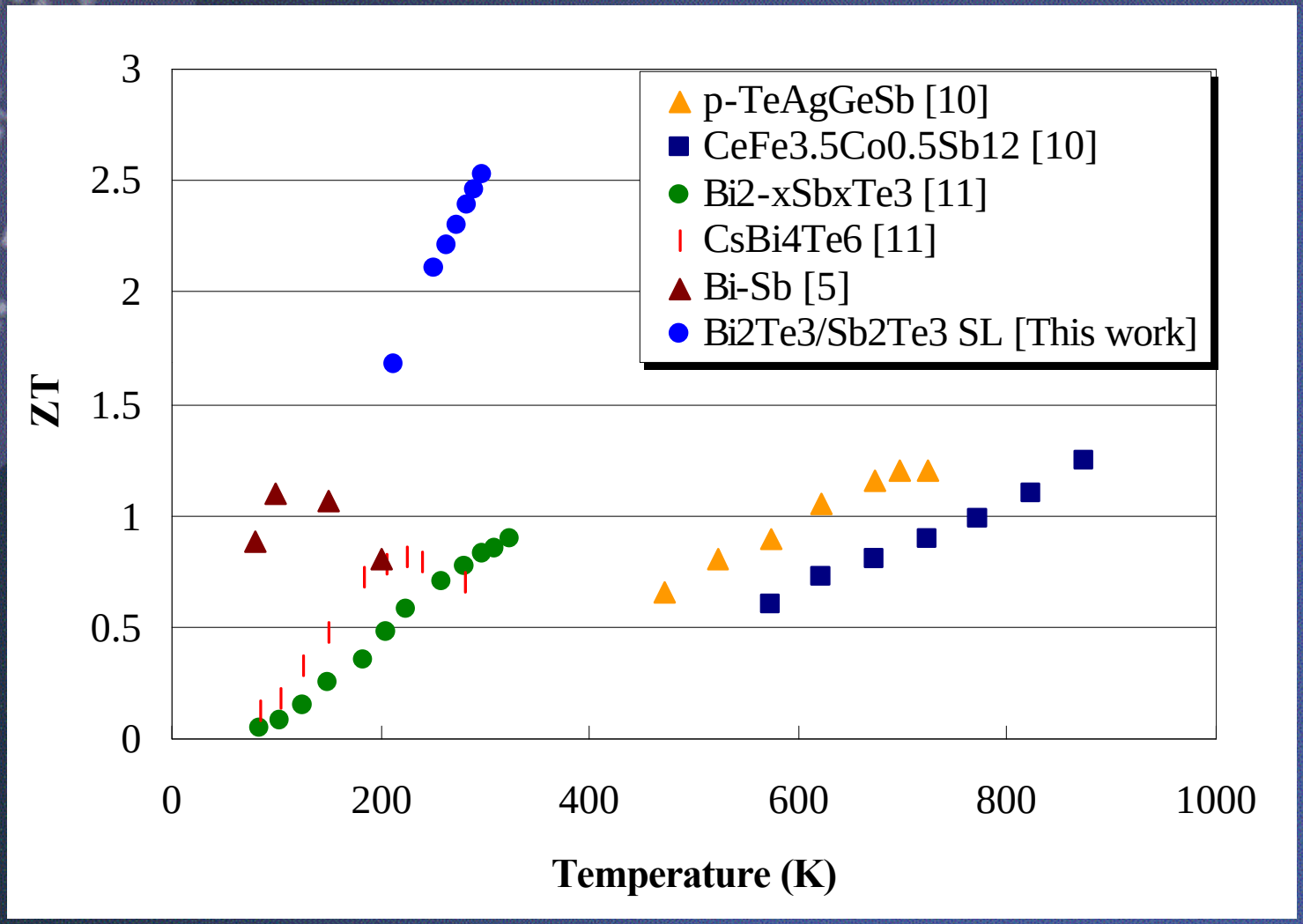
$$ZT_i = [\partial V_0/\partial (I.l/a)]/[\partial V_R/\partial (I.l/a)]$$

ZT by Variable-Thickness Method in Bulk Materials Thinned Down 5 to 20 microns

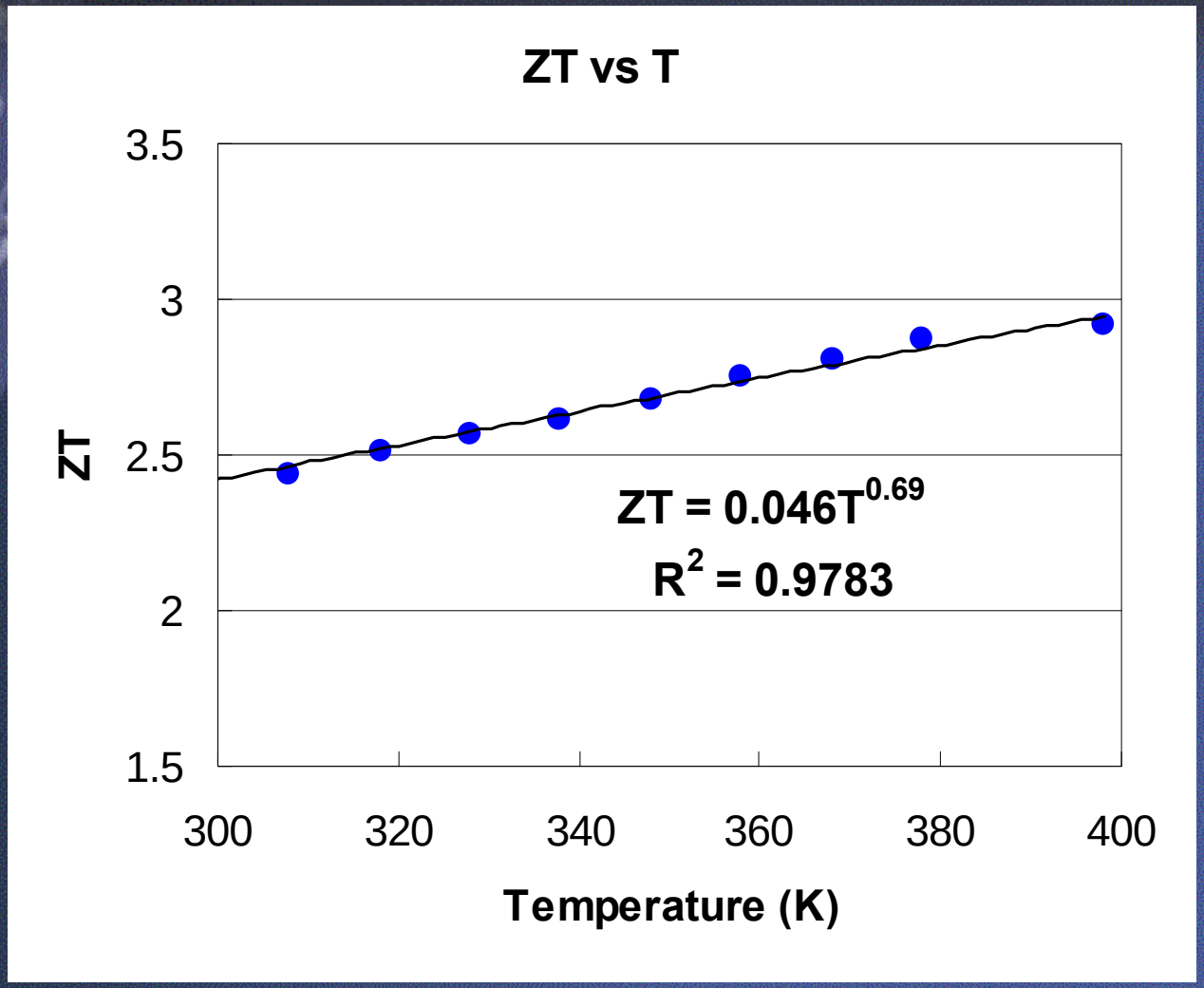


$ZT_{intrinsic} \sim 1.1$

Temperature dependence of ZT in p -type Bi_2Te_3/Sb_2Te_3 Superlattices (213-300K)

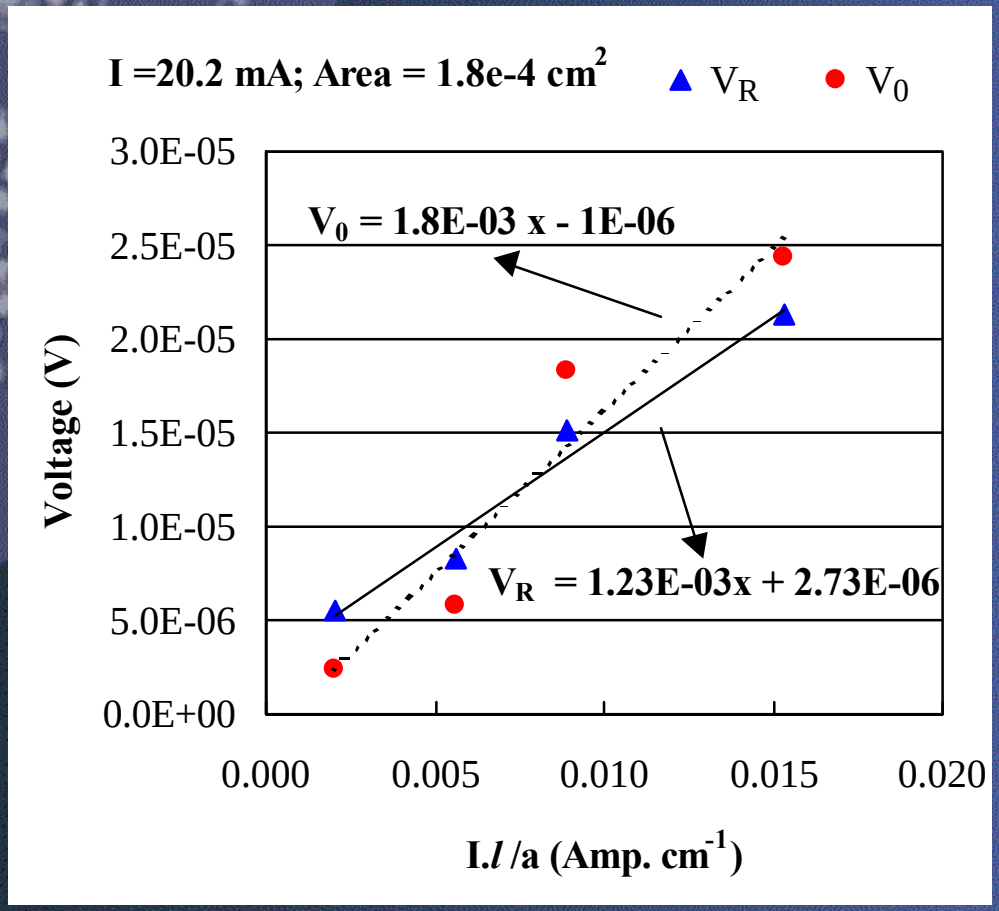


Temperature dependence of ZT in p -type Bi_2Te_3/Sb_2Te_3 Superlattices (300-400K)



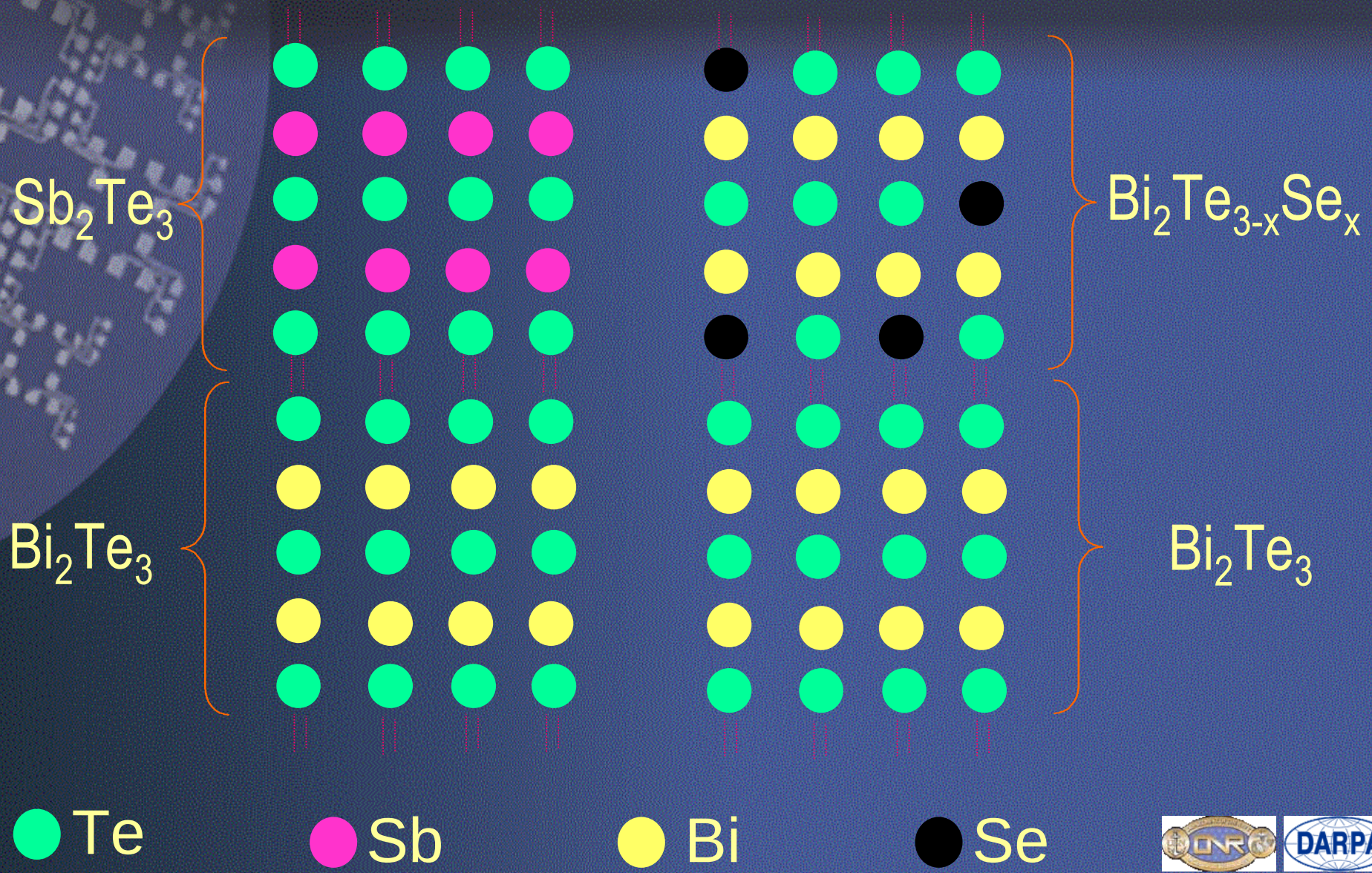
- Ref: DARPA Palm-Power Program, Dr. Robert Nowak

ZT in n -type $Bi_2Te_3/Bi_2Te_{3-x}Se_x$ Superlattices by Variable-Thickness Method

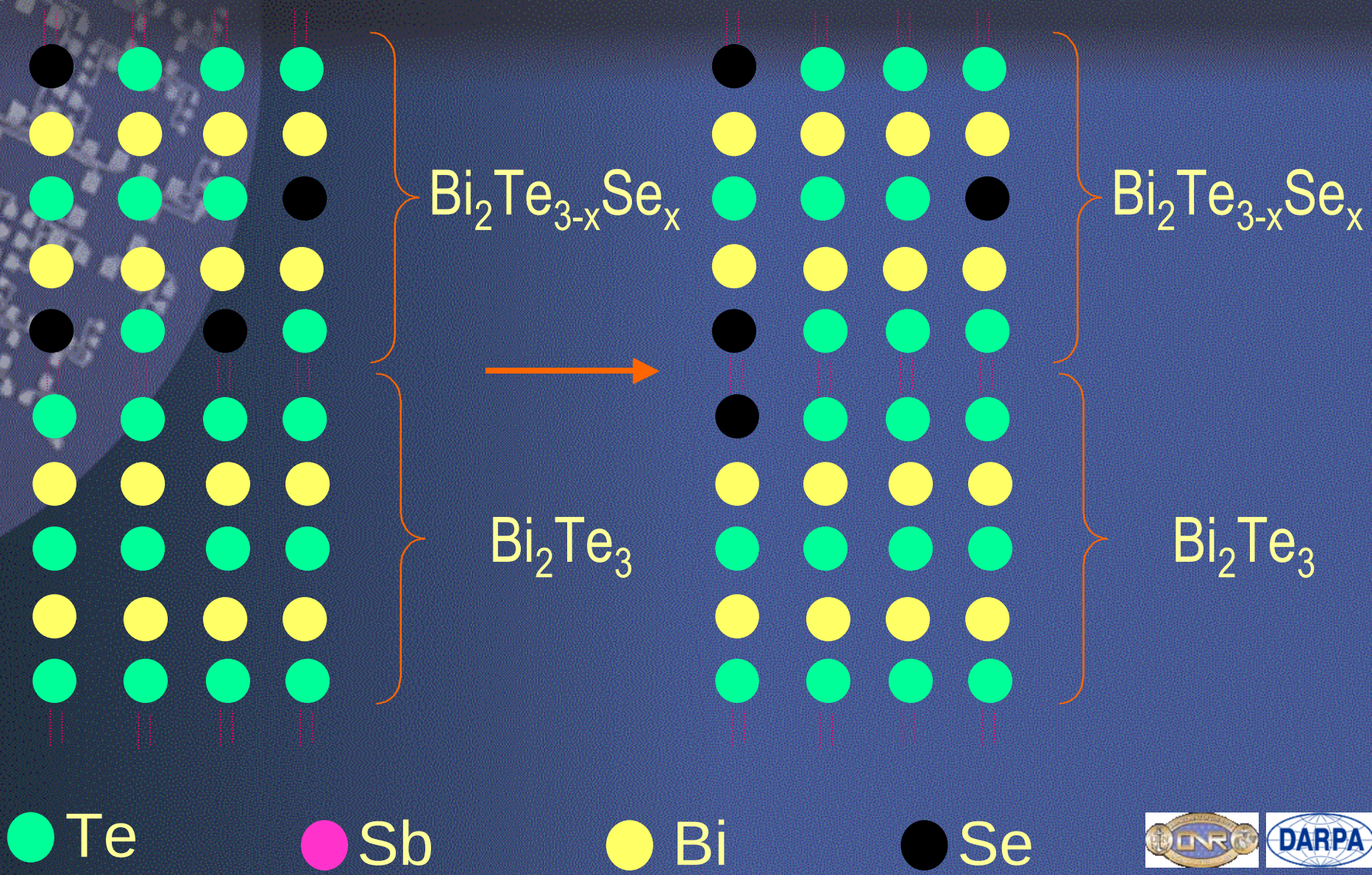


- **$ZT_{intrinsic}$ ~ 1. 4**
- **Limited by $K_{Lattice}$ ~ 5.8 mW/cm-K compared to ~2.5 mW/cm-K in p-type Bi_2Te_3/Sb_2Te_3 superlattices**
- **Even so, ZT along c-axis three times better than in bulk**

Comparison of p-type Bi_2Te_3/Sb_2Te_3 superlattices and n-type $Bi_2Te_3/Bi_2Te_{3-x}Se_x$ Superlattices



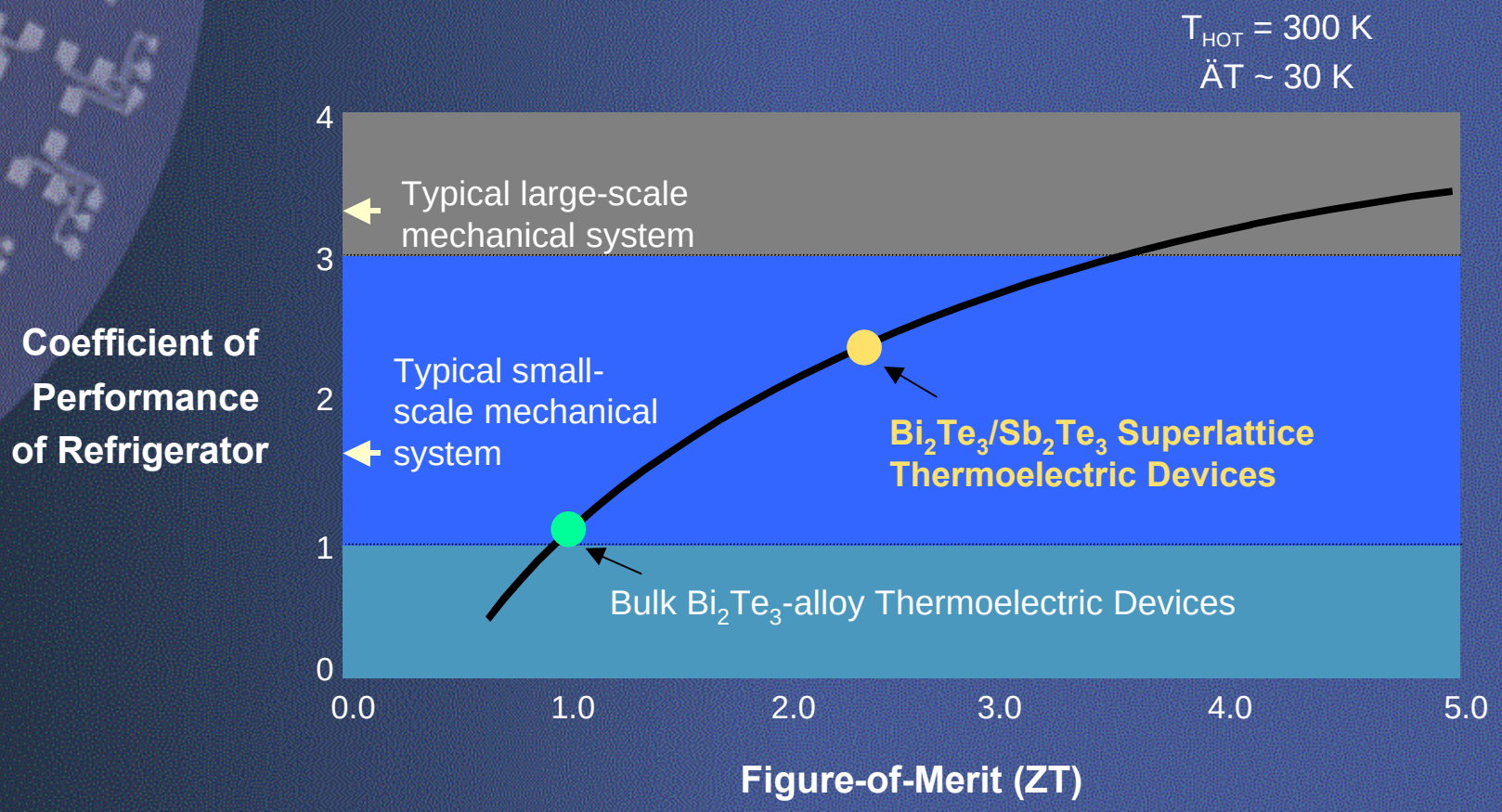
Potential for non - ideal n -type $Bi_2Te_3/Bi_2Te_{3-x}Se_x$ Superlattices



Comparison of p-type Bi_2Te_3/Sb_2Te_3 superlattices and n -type $Bi_2Te_3/Bi_2Te_{3-x}Se_x$ Superlattices

- p-type Bi_2Te_3/Sb_2Te_3 superlattices
  - **"ideal " components – <u>hand-shake of phonons from one layer to other difficult</u>**
    - **Lattice thermal conductivitv falls to 2.5 mW/cm-K**
- n -type $Bi_2Te_3/Bi_2Te_{3-x}Se_x$ Superlattices
  - **"non-ideal " components with perhaps some inter-layer mixing – <u>hand-shake of phonons from one layer to other relatively easy</u>**
    - **Lattice thermal conductivitv does not fall; in fact closer to that of alloy along c-axis (5.8 mW/cm-K)**

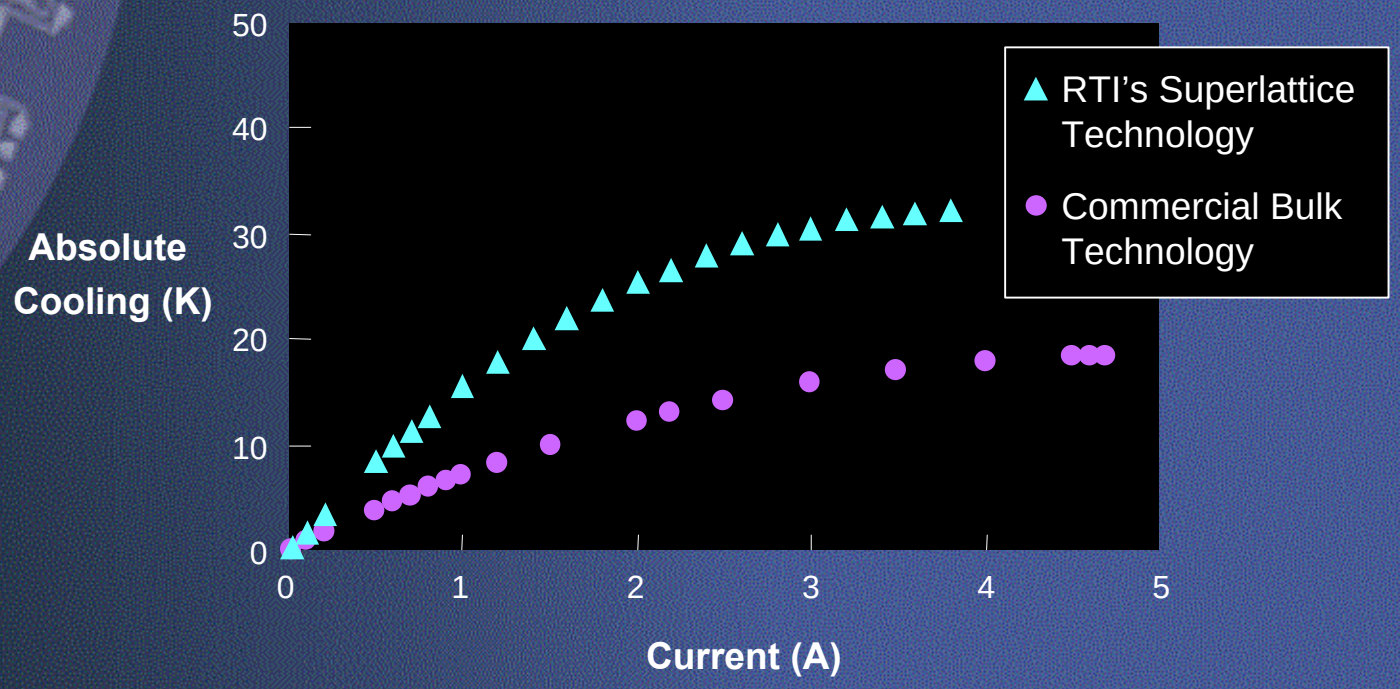
RTI's Superlattice Device Improves Refrigeration Efficiency



Higher ZT – Incentives for New Approaches to Implement Higher COP Concepts

Advantages of RTI's Superlattice Thermoelectric Technology

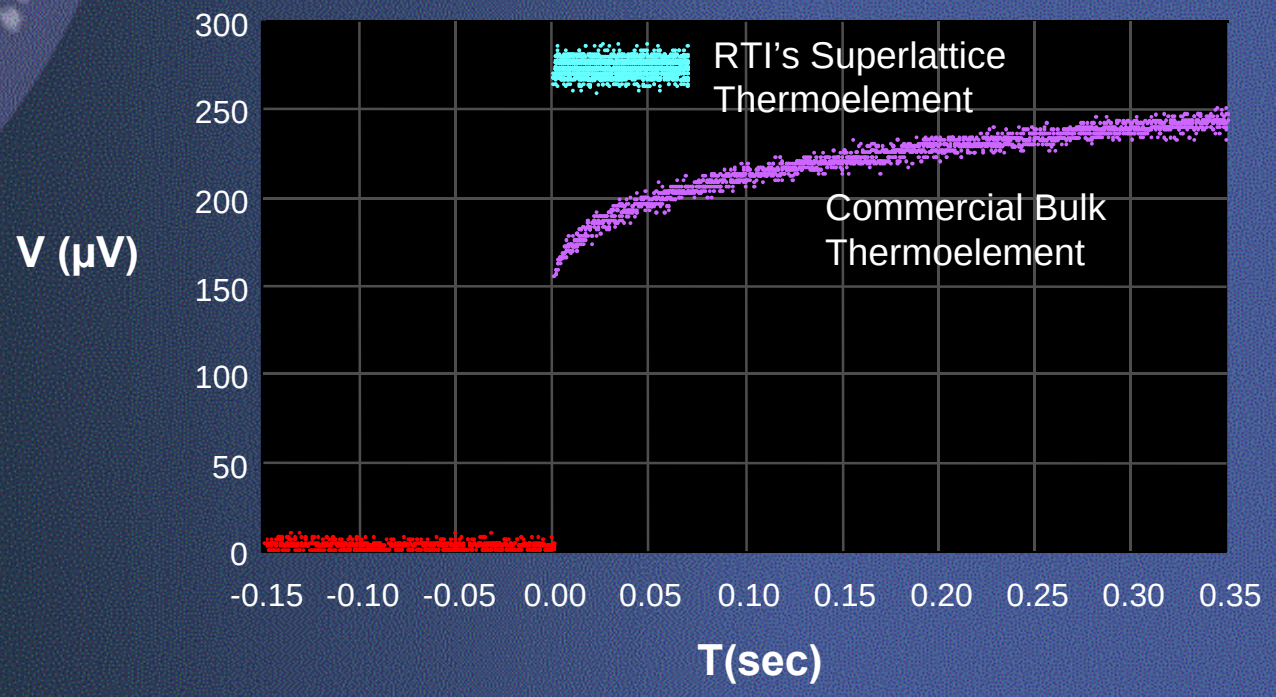
- Enhanced cooling



Other thin-film technologies have one-tenth the cooling of the RTI superlattice devices

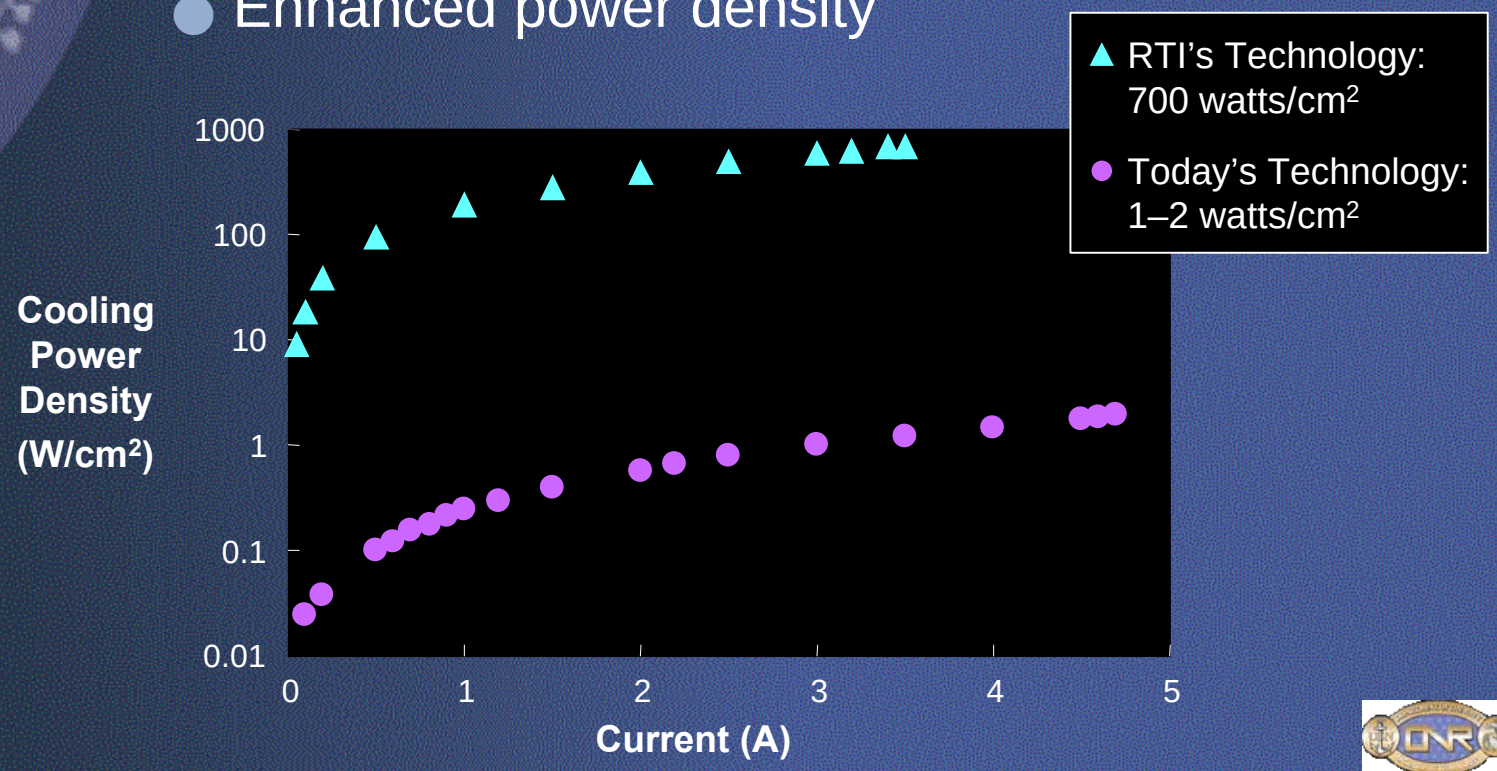
Advantages of RTI's Superlattice Thermoelectric Technology

- Enhanced cooling
- Super-fast cooling and heating



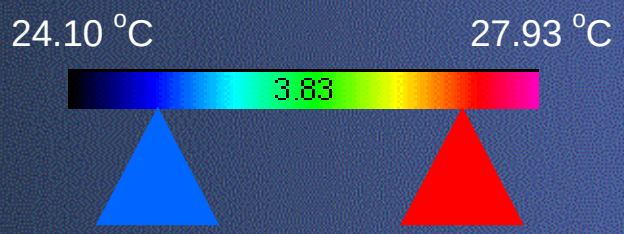
Advantages of RTI's Superlattice Thermoelectric Technology

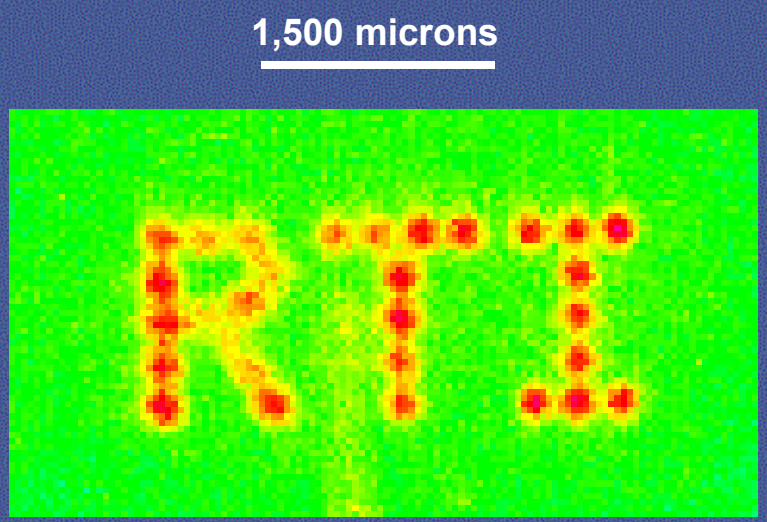
- Enhanced cooling
- Super-fast cooling and heating
- Enhanced power density



Advantages of RTI's Superlattice Thermoelectric Technology

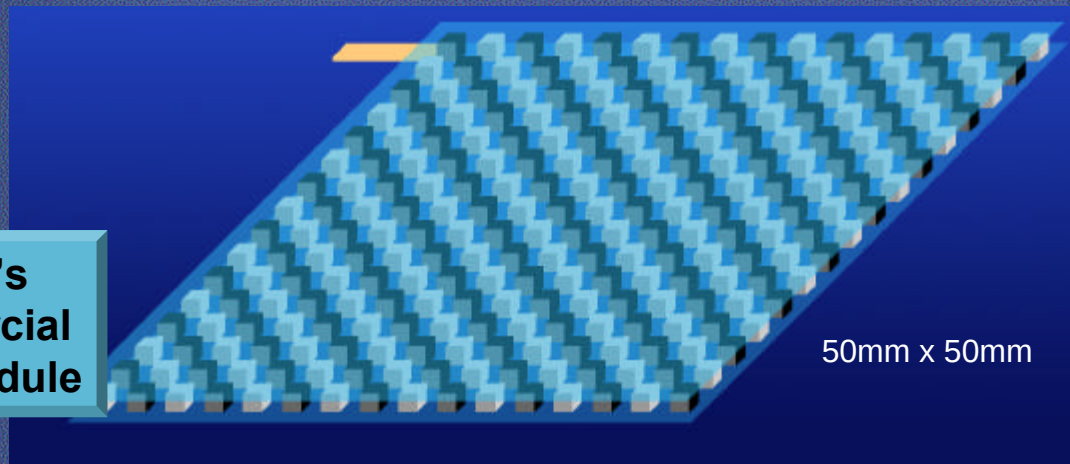
- Enhanced cooling
- Super-fast cooling and heating
- Enhanced power density
- Anywhere, any time cooling/ heating technology





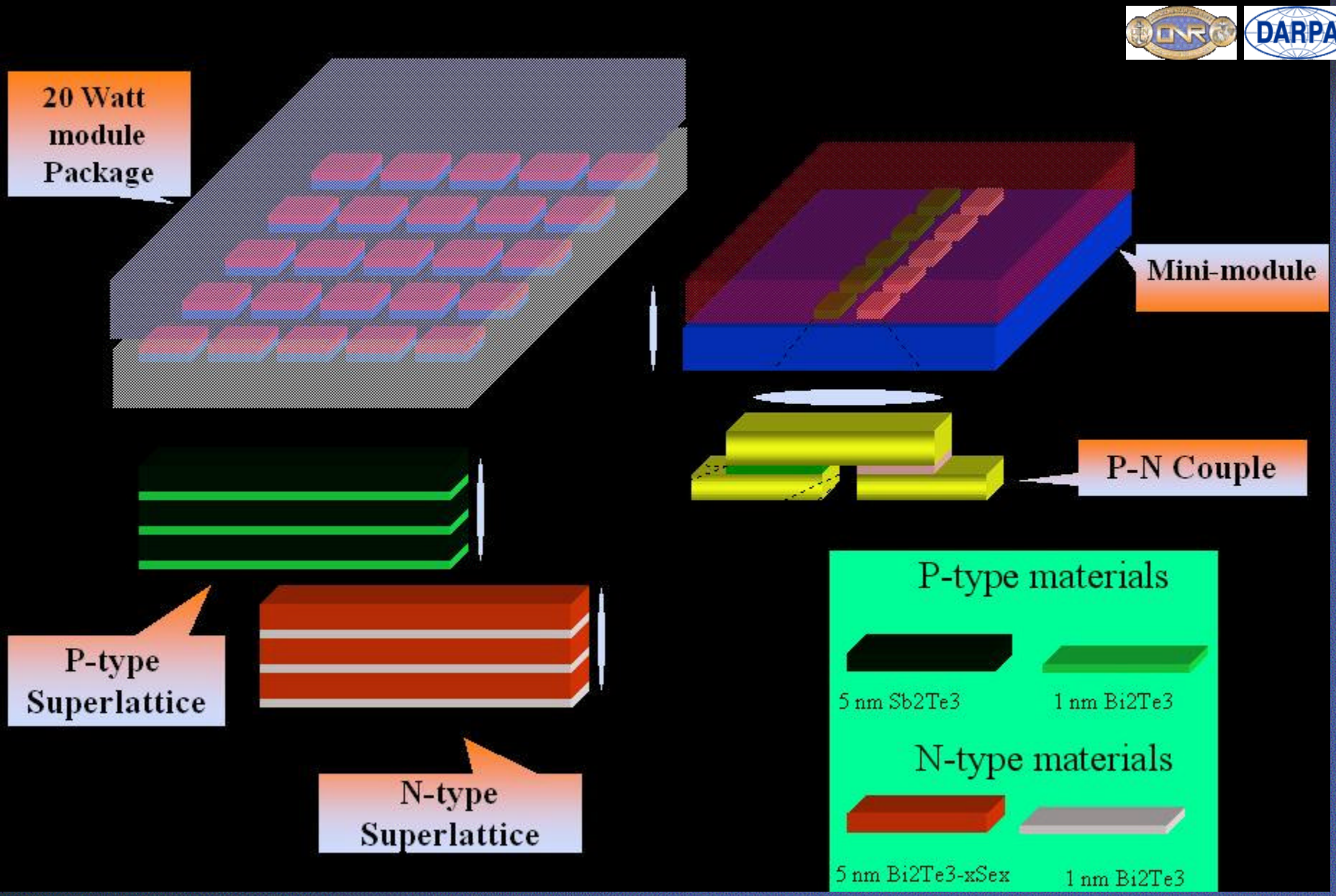
Advantages of RTI's Superlattice Thermoelectric Technology

- Enhanced cooling
- Super-fast cooling and heating
- Enhanced power density
- Anywhere, any time cooling/ heating technology
- Significantly more compact



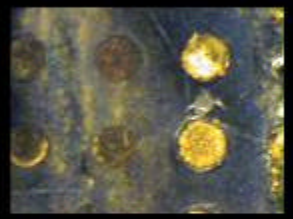
RTI’s Progress in Cooling Module Development

Thin-film Thermoelectric Module Fabrication



Progress in RTI's Superlattice Thermoelectric Module Technology

150 μm



Single p-type-element

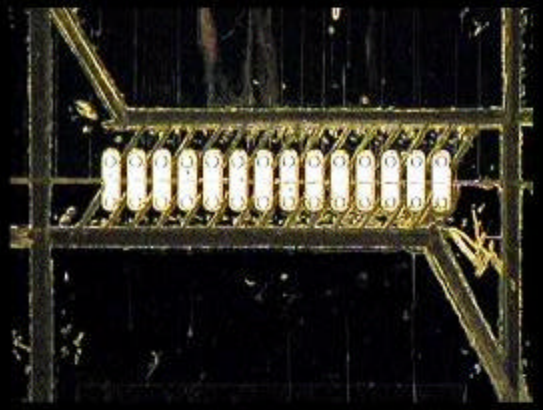
32K Cooling

(Ref: *Nature*)

300 μm

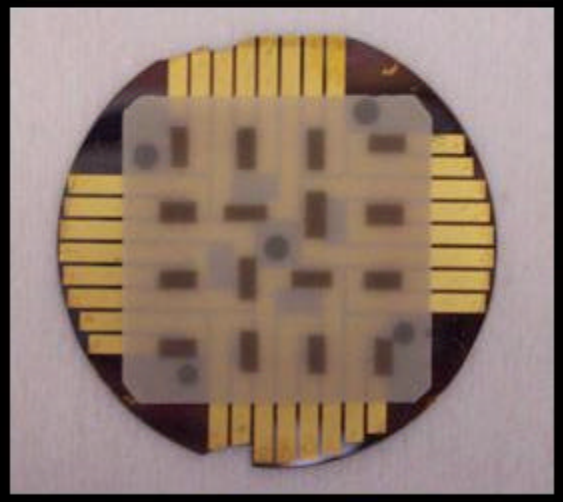
P-N Couple

500 μm



14 P-N Couples Series Connected Mini-module

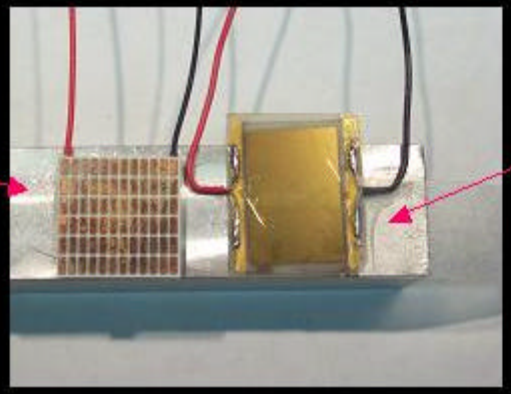
6000 μm



Wafer-Scale Integrated Module Composed of 16 mini-modules

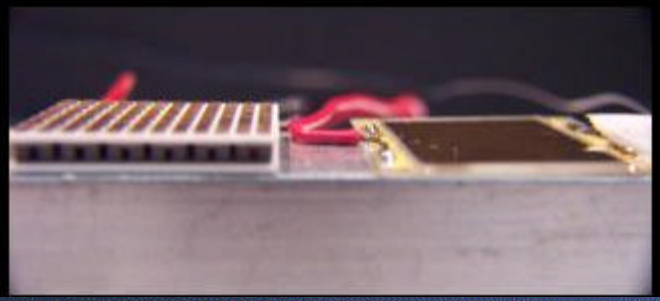
Comparison of Commercial Bulk and Prototype Thin-Film Modules (that we are beginning to ship for early co - development customers)

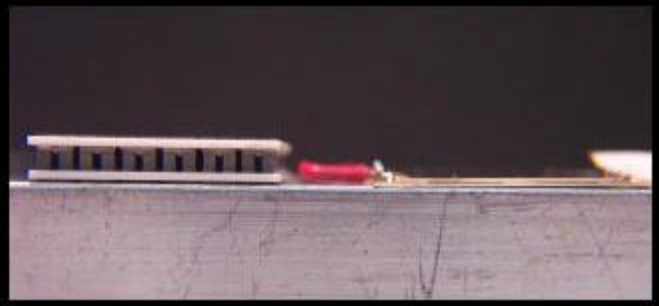
Bulk Module



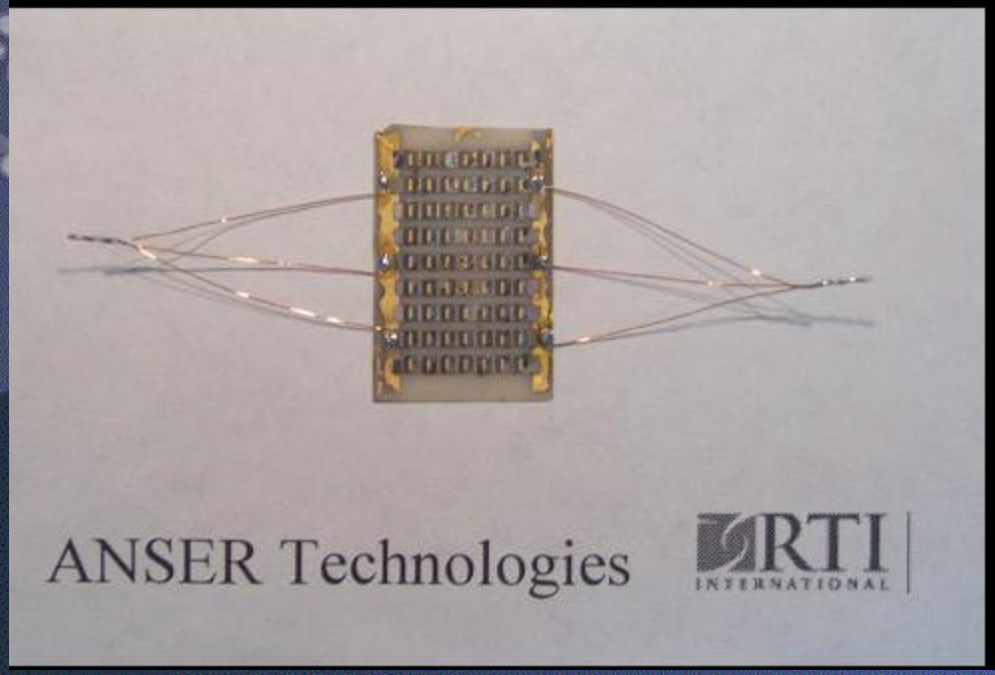
Thin-film Module (2 cm x 2 cm)

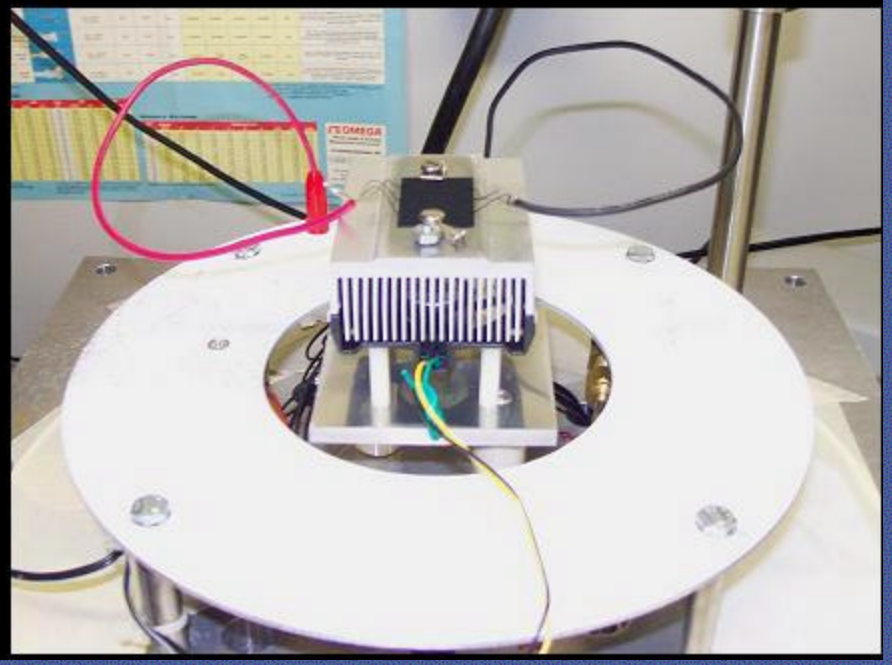
Profile Comparison (Bulk vs Thin-film) of Completely Packaged Modules





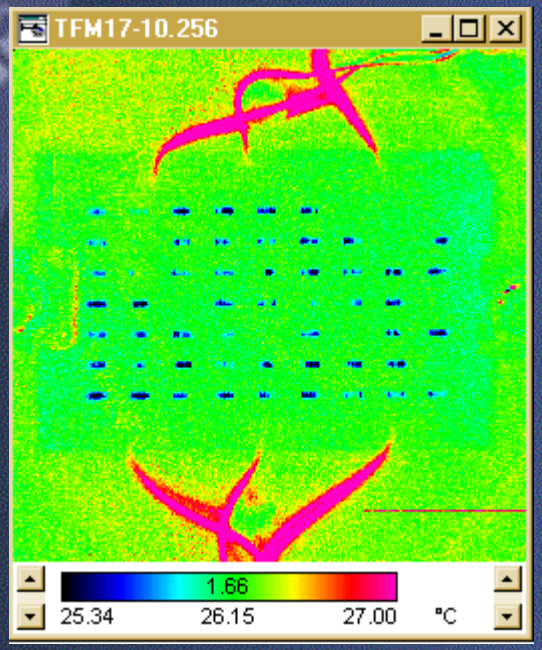
3.5 cm x 2.4 cm Thin-film Module that can be inserted on top of a commercial heat–sink to which a high-speed microprocessor can be attached



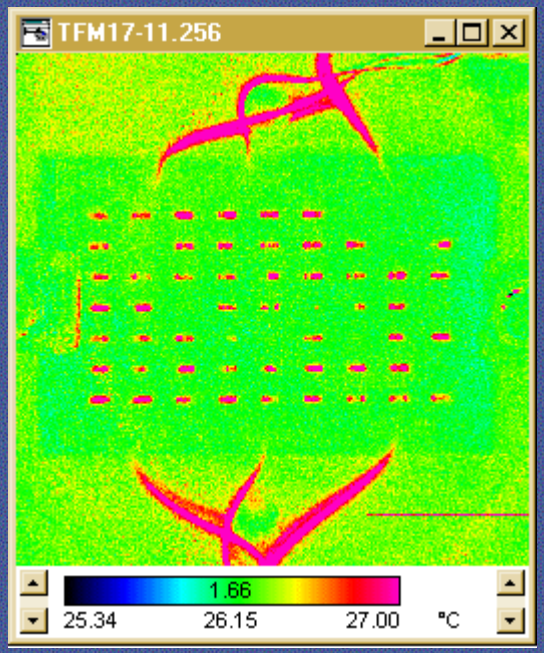


IR Images From a 630-element TE Module; 630 elements arranged in a 9x7 array of mini-modules; each mini-module has 5 p-n couples

Cooling



Heating

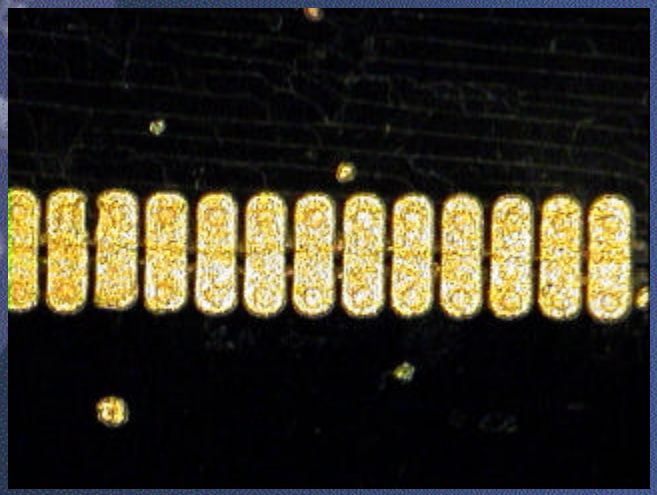


35 mm

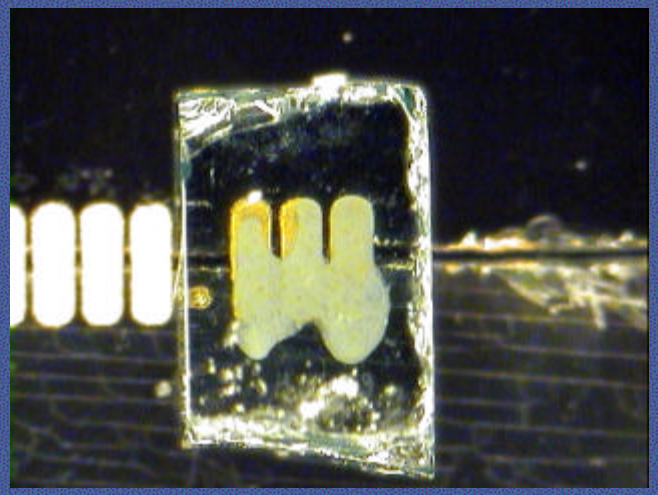
- Note only ~ 90 mA through each element
- Number of elements typical for a bulk module ~ 256

High-Active Flux, Low Input-Output Flux (HAF-LIOF) Concept for Large-Scale Cooling Modules

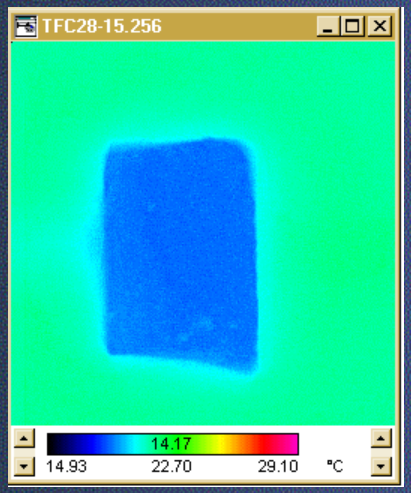
A 2 x 14 element or 14-couple wafer



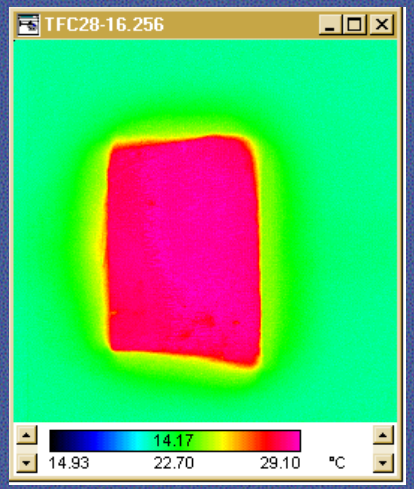
A 2 x 3 element or 3-couple module with a High-l header



Cooling of whole header due to thermal spreading effect



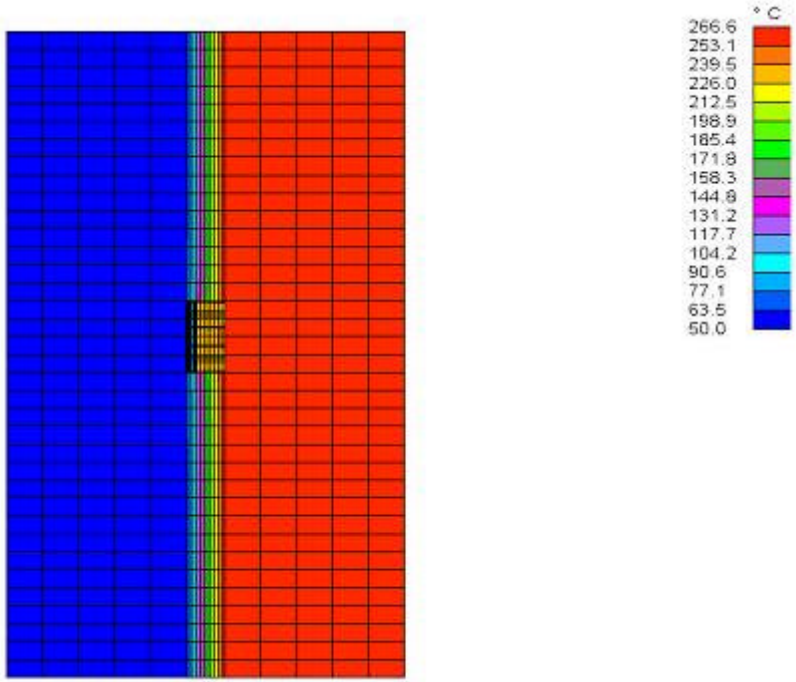
Heating of whole header due to thermal spreading effect



Thermal Modeling Studies

- Windows based general purpose thermal modeling software
- Finite Element Approach – linking algebraic equations together
- Built in steady-state and transient finite difference solvers
- All modes of heat transfer included
  - Conduction
  - Convection
  - Fluid Flow
  - Radiation
- All properties can be defined as time and/or temperature dependent
- Size of model/speed of solution is hardware limited

Thermal Modeling of High-Active Flux, Low Input-Output Flux (HAF-LIOF) Concept



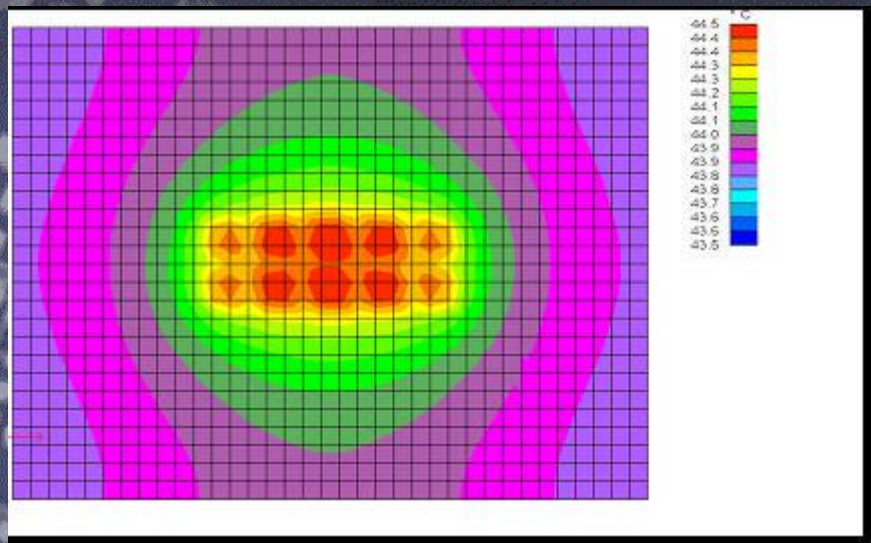
- Example:>1500 W/cm^2 through elements
- 1% coverage of TE elements
- ~15 W/cm^2 at the heat source or heat sink

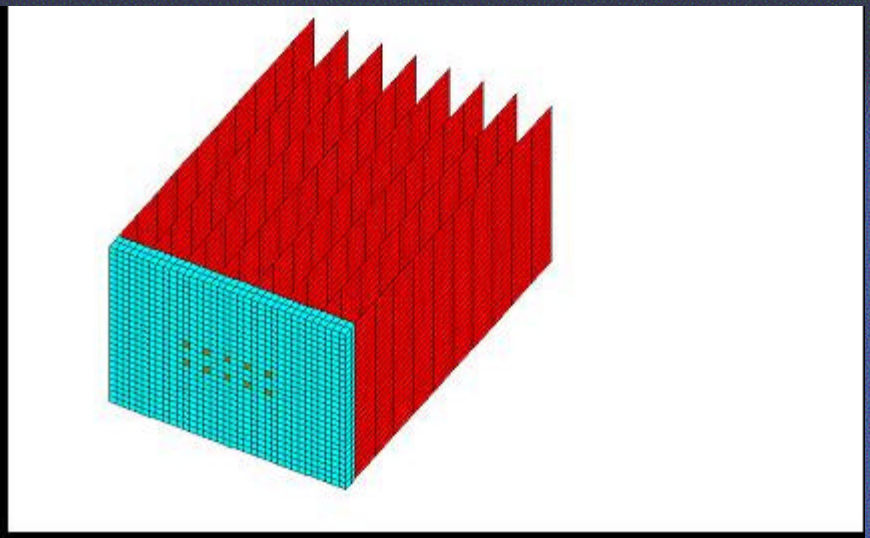
- Enormous Implications for TE Material Requirements for Increased Applications, with Higher Efficiencies that can be Achieved Now

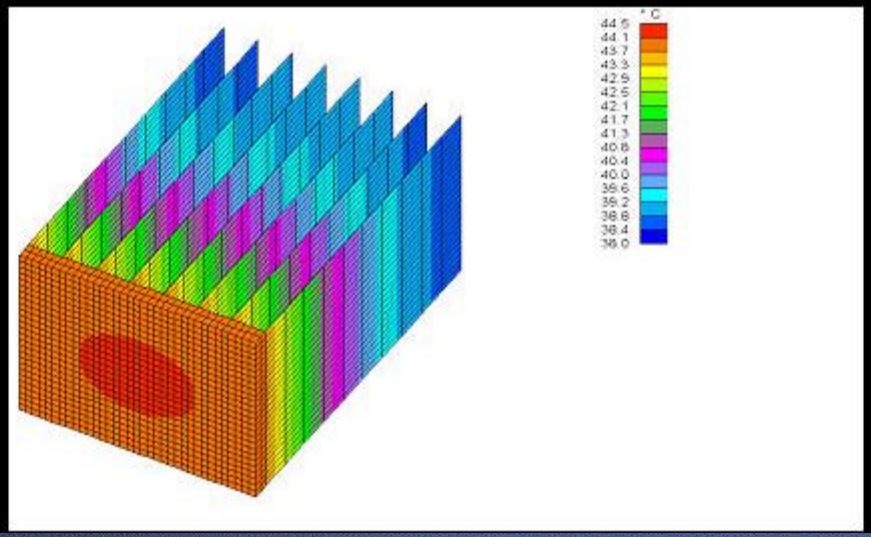
Progress in Heat-Rejection System Development for Large-Areas

- High Active Flux, Low Input-Output Flux (HAF-LIOF) Devices should reduce heat-rejection concerns
  - **Low packing fraction of active thermoelectric devices and high-thermal conductivity heat-spreaders so that excessive heat fluxes are not required at source and therefore not dissipated at sink**
  - Minimizes amount of thermoelectric materials required and larger efficiency from higher active-ΔT in power generation
- Working with commercially available sources of high-efficiency, light-weight, heat rejection system design and prototyping
- In-house modeling and fabrication capability

Modeling Heat Pumping to Heat Rejection







Program Plans in the Cooling Program Area

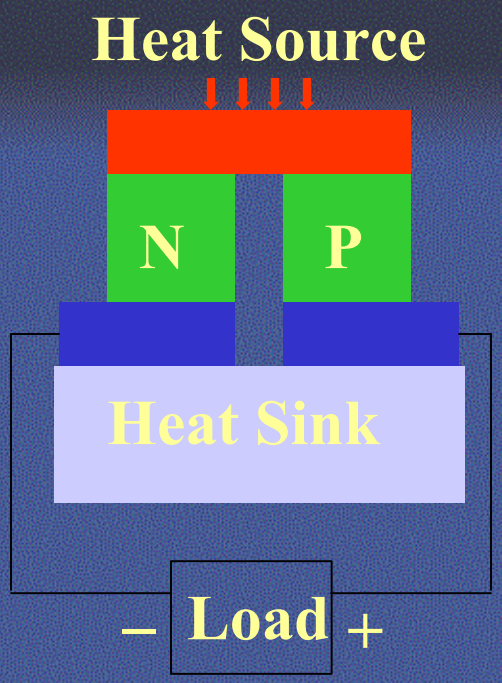
- **Materials Development**
  - Improved N-type Materials to achieve an average ZT of ~2.5 for the p-n combination
  - Develop films with larger effective thickness to minimize thermal management problems
- **Device and Module Development**
  - Identify Approaches to reduce internal thermal and electrical resistances in devices and modules to recover COP consistent with higher ZT
- **Heat Rejection System Development**
  - Demonstrate design feasibility to remove ~100 Watts of heat over 4 to 6 cm^2
- **Preliminary validation of integration with applications in spot cooling, chip cooling, optoelectronics cooling**

RTI’s Progress in Power Devices

(Ref: DARPA Palm-Power Program; Dr. Robert Nowak)

ZT, ΔT and Power Conversion Efficiency

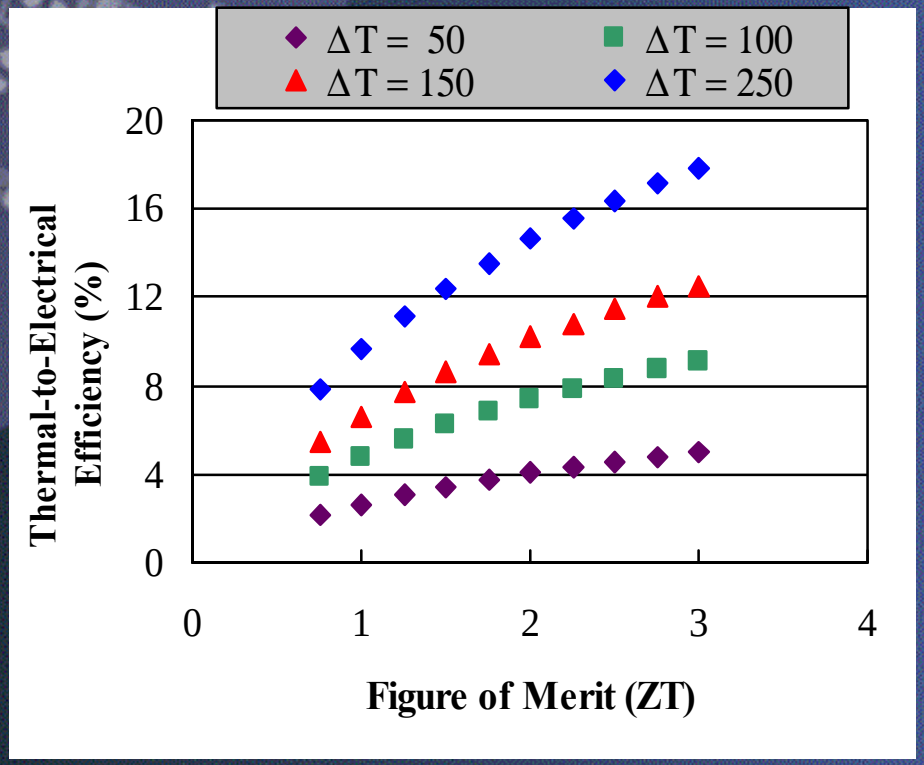
- Need high ZT in both p and n-type materials
- In addition to high ZT, need higher DT or T_h-T_c for higher efficiencies
- Thermal management (getting the heat out from the heat-sink) is important to generating the maximum DT

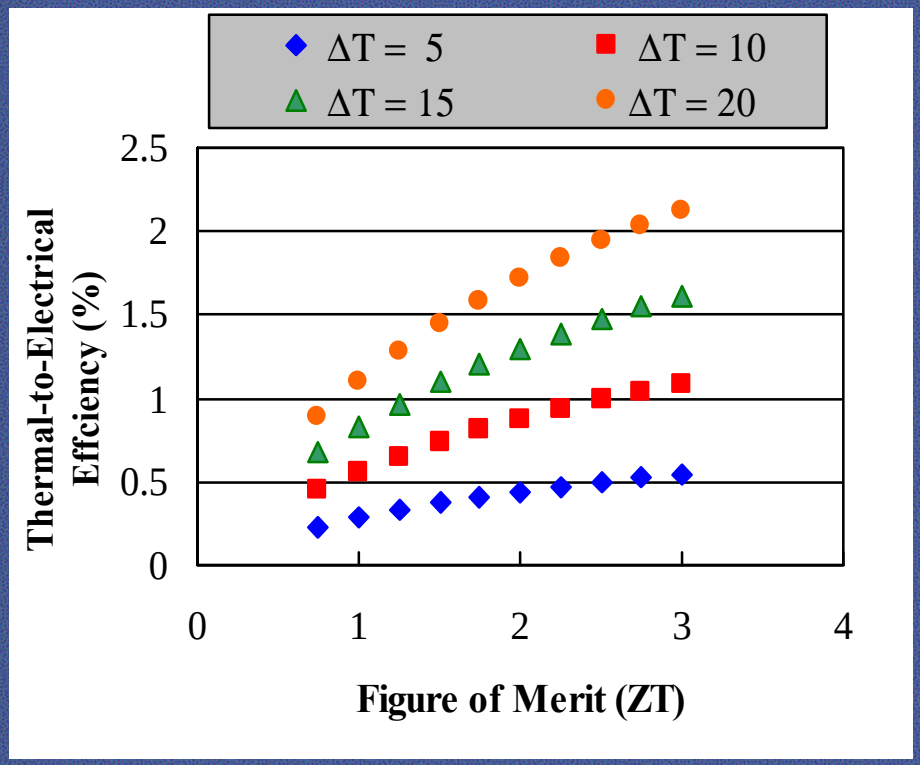


$$y = \frac{(T_h - T_c)}{T_h} \; \frac{\{(1+ZT)^{1/2} - 1\}}{\{(1+ZT)^{1/2} - 1\} + T_c/T_h}$$

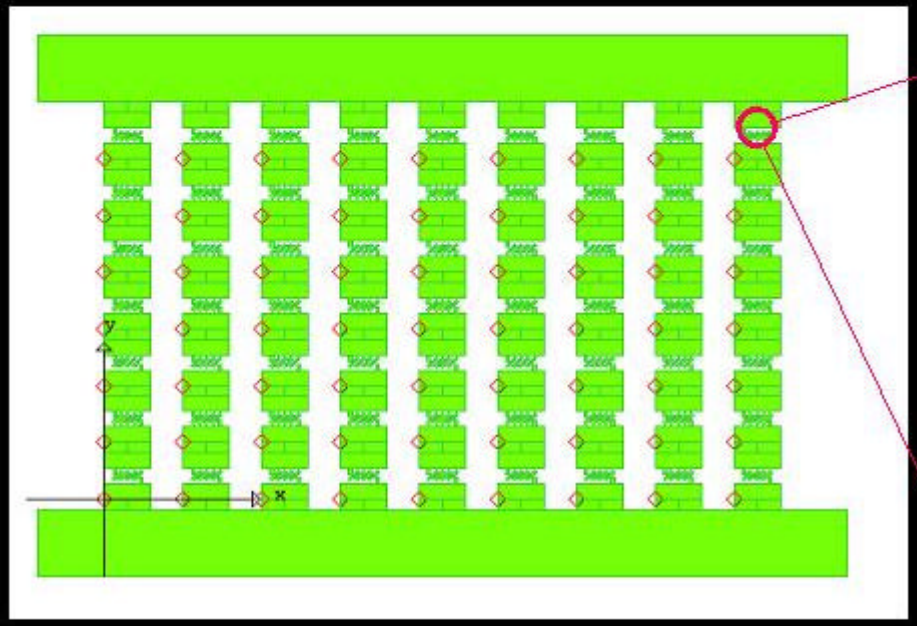
Implications for Power

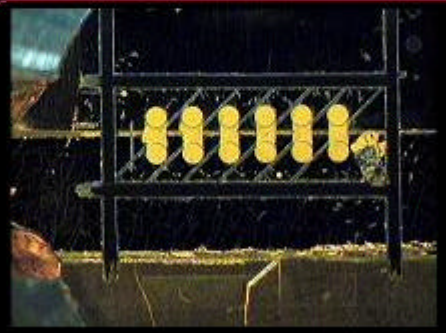
- High-density (area, volume, weight) power
- Higher efficiency at low ΔT





RTI's Gen-1 Power Module Design Using 63 Mini-modules (Area ~ 3.5 cm x 2.4 cm)

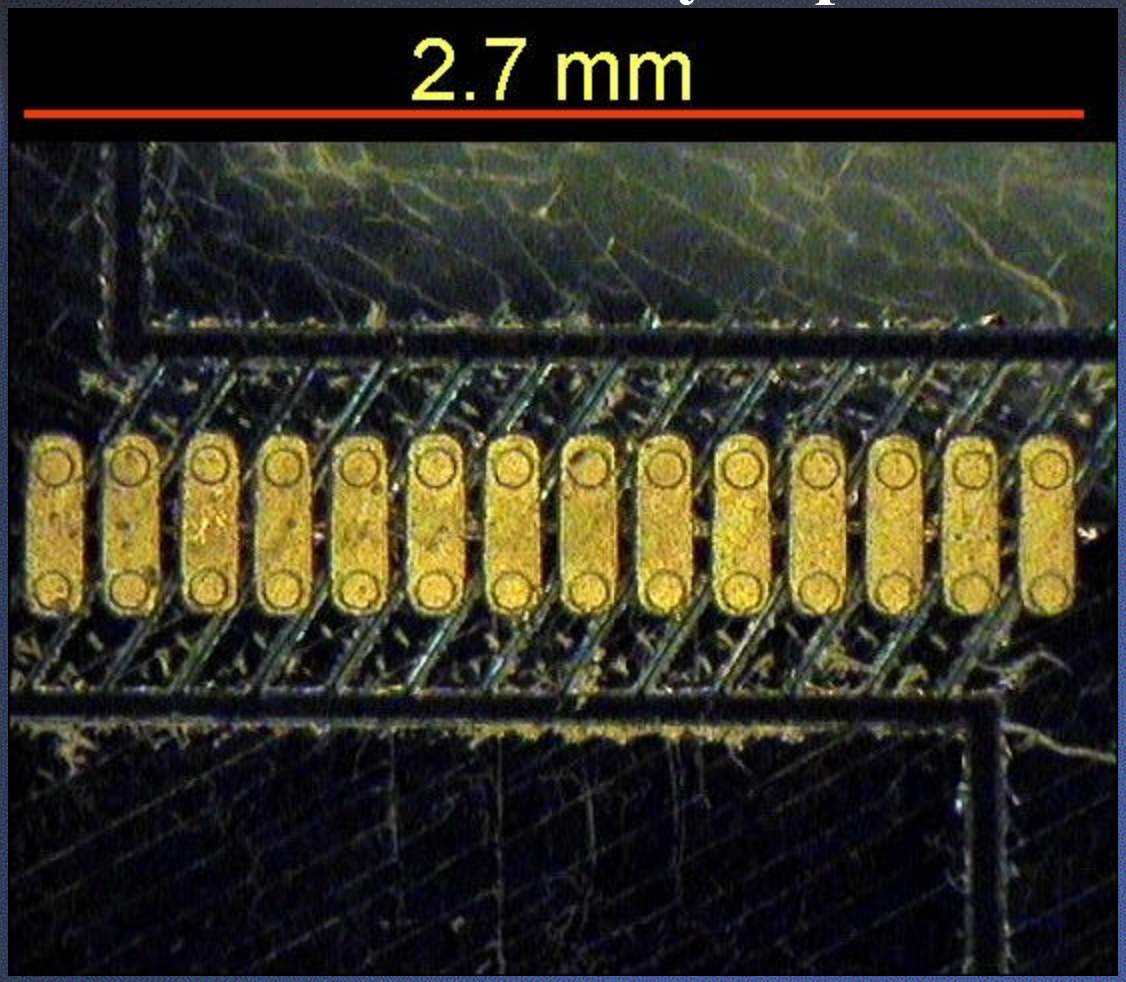




- **6x2 300-μm Series mini-module**
- **V_{oc} ~ 76 mV**
- **P_{max} = 3.160 mW**
- **ΔT ~ 25°C**

- One year target of 5 Watts Module based on power from all 63 mini-modules, ΔT of 75°C, factor of three improvement in reducing internal series resistance and translating current material ZT to external ZT
  - 3.16 mW x 63 x $(75/25)^2$ x 3 ~ 5 W
- **Larger Immediate Powers Possible with Larger Area Modules for Immediate Applications as in RTI Thermoelectric Power Shields™**
- Three-year target of 20 Watt Based on ΔT of 150°C

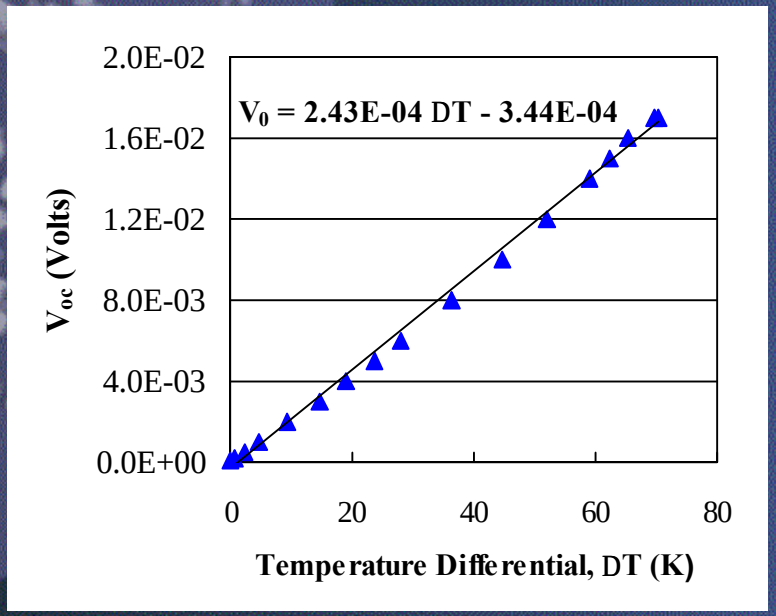
28 element mini-module: all elements electrically in series and thermally in parallel



- Larger Number of Element Based Mini-module for larger voltages

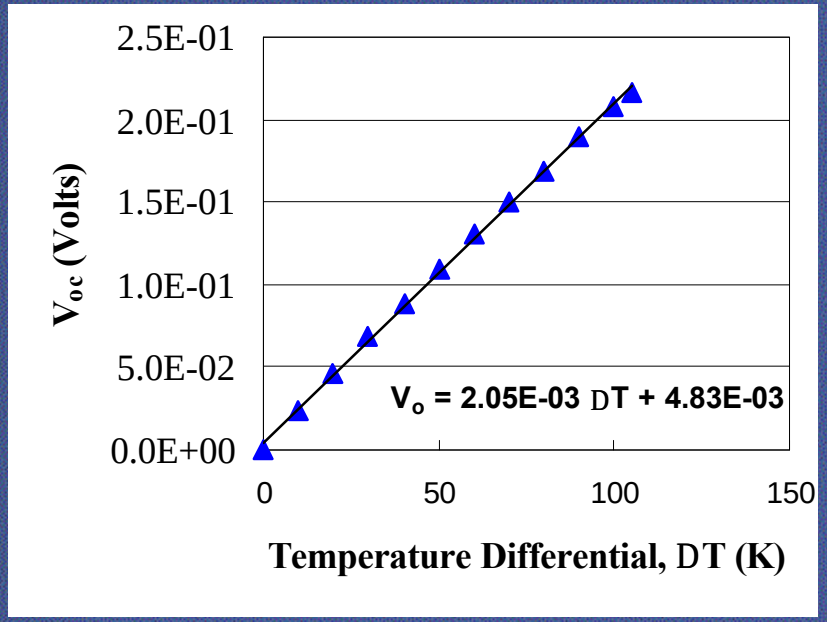
V_{OC} Progress

Palm-Power Program Start



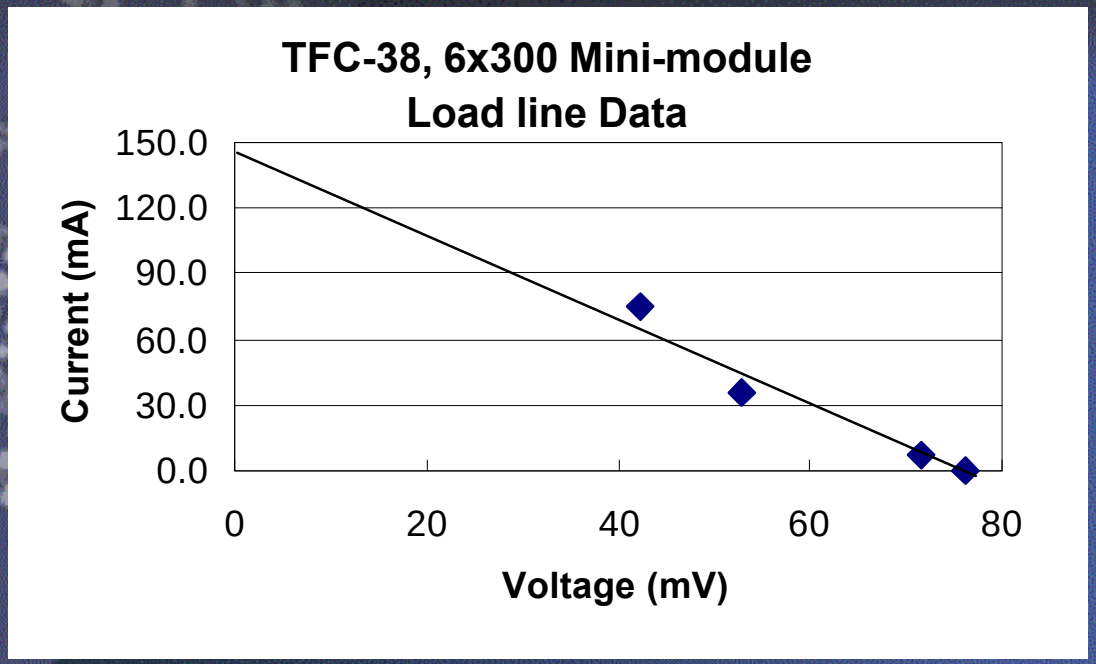
- 1-element
- V_{oc} ~ 17 mV

As of Now



- 28-element mini-module
- V_{oc} ~ 220 mV
- Target milestone of 200 mV achieved

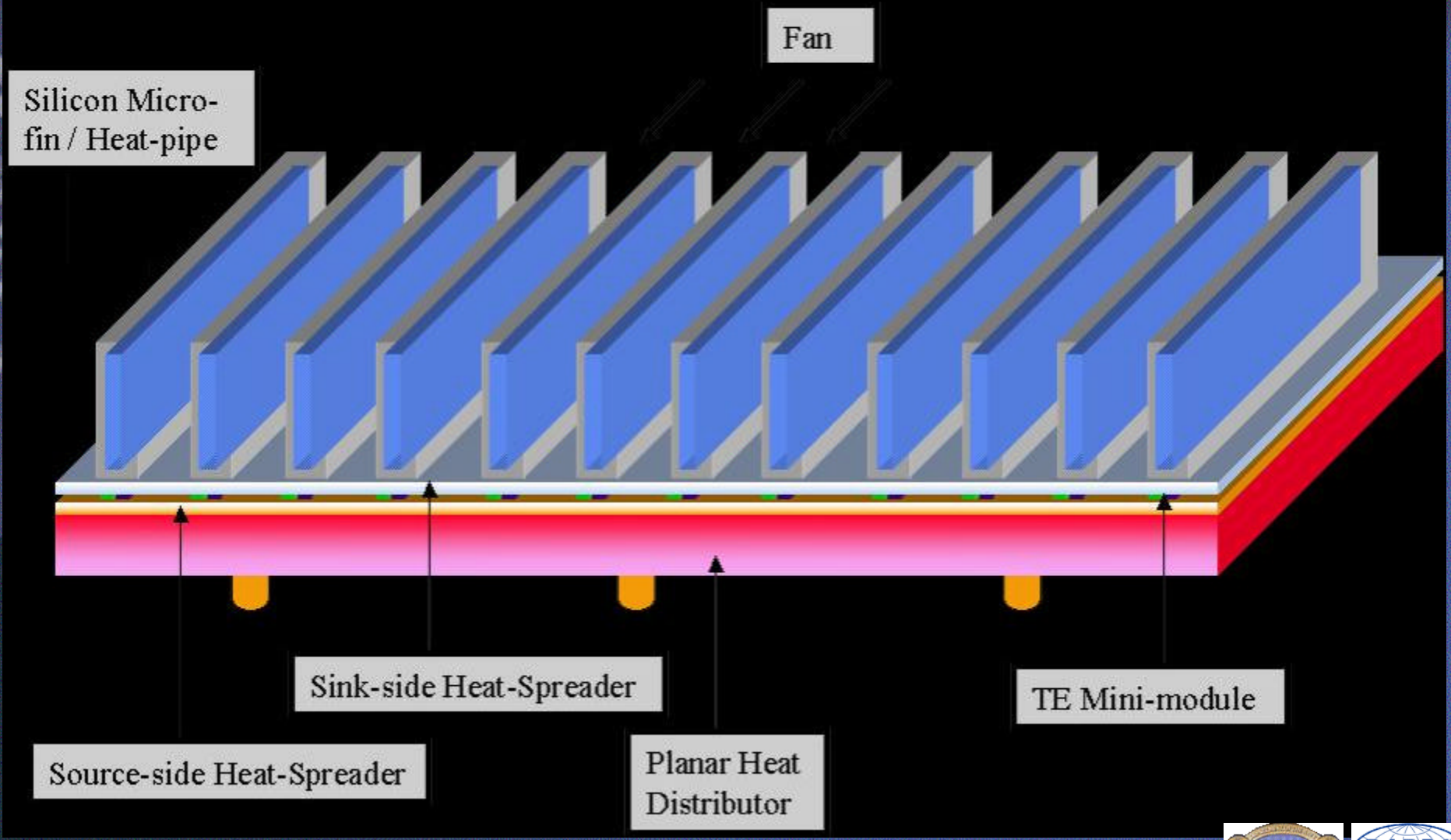
Typical load line data from a thin-film mini-module as of today - early devices



- 12-element mini-module
- V_{oc} ~ 76 mV
- Potential I_{sc} if no internal resistance ~ 140 mA
- At peak power of 3.16 mW
  - ΔT_{device} ~ 25K
  - V_{load} ~ 42. 2 mV
  - I_{load} ~ 74.9 mA
  - Typical power output from non-superlattice commercial thin-film device for same ΔT ~ 25 μW

- J_{sc} of this mini-module ~ 3.3 A/cm^2
  - Typical <u>bulk module</u> for this ΔT, J_{sc} ~16 mA/cm^2

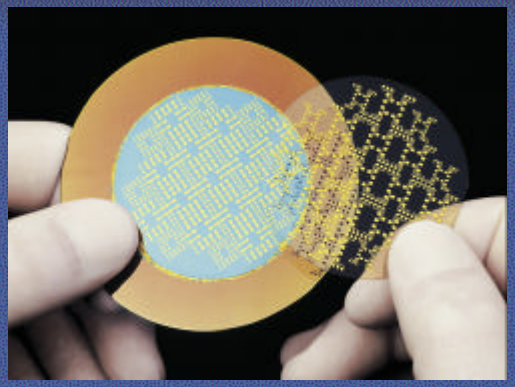
Heat-Source / Heat-Spreader / TE Module / Heat Spreader / Integrated Microfin-Heat Pipe Heat Exchanger



Emerging Applications

Application in Genomics and Proteomics

- High-speed PCR for rapid DNA analysis
- Self-assembly of DNA molecules
- Today's analytical tools are inadequate
  to study proteins
  (2-D gel electrophoresis, liquid chromatography, and mass spectrometry)
- Wafer-scale thermoelectrics can enable "fingerprinting" of proteins and their interactions

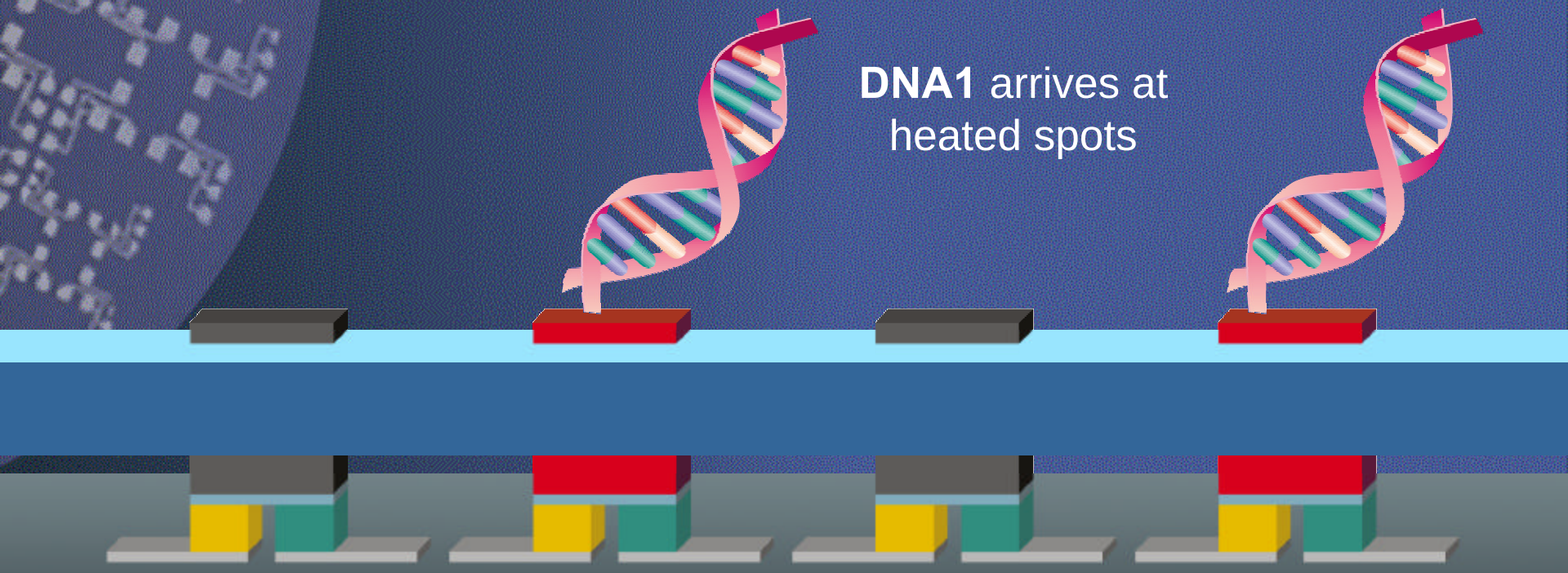


High-speed PCR

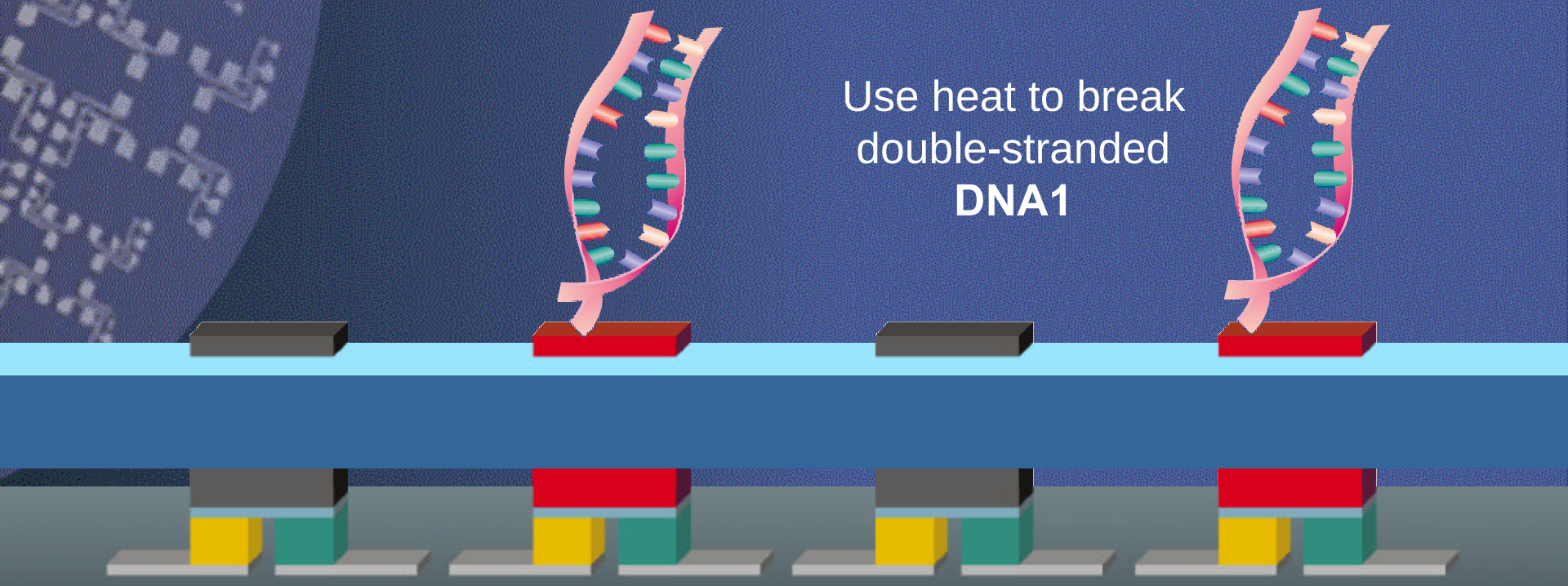
- Typical PCR- starts with double-strand DNA being split at 95°C, temperature lowered to where single strands can bind together with new strands
  - Two-step heating cycle is repeated n times, produces 2^{n} copies of target sequence
  - From a single starting molecule, 20 cycles yield over 1M, 30 cycles 1B, and 40 cycles 1T copies
  - Typically each cycle is 1 minute
- Reducing cycling time, efficient devices, and small-scale battery-operated systems can lead to field applications for DNA testing for pharmacology and bio-weapon detection

Self-Assembly of DNA Microarrays

Self-Assembly of DNA Microarrays

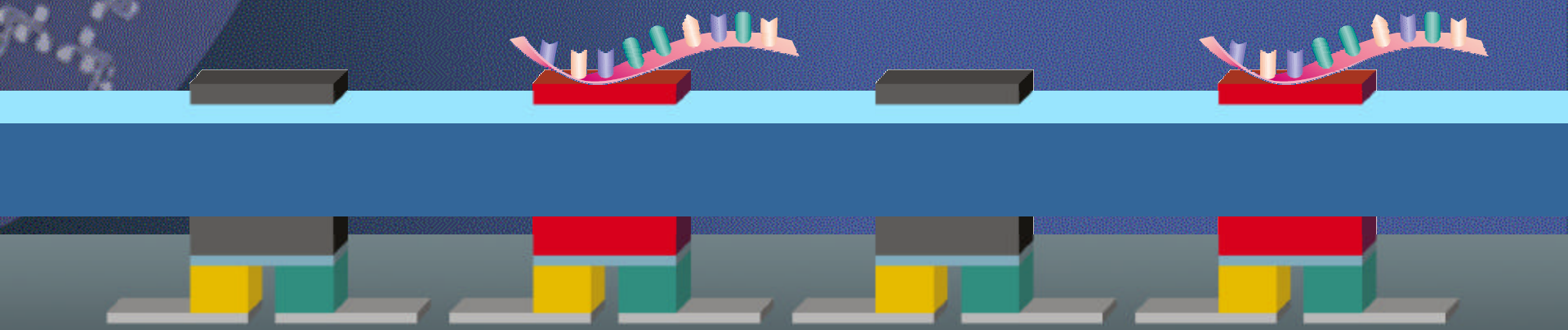


Self-Assembly of DNA Microarrays



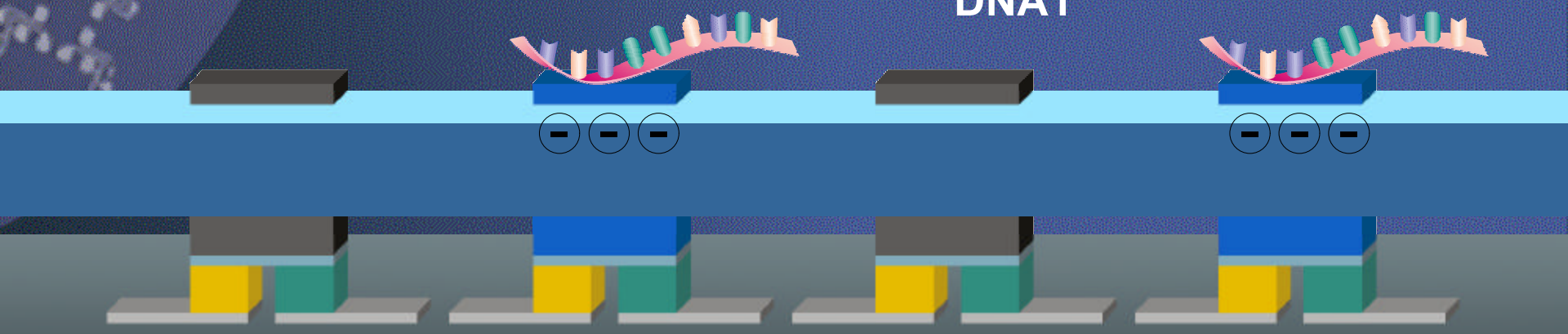
Self-Assembly of DNA Microarrays

A single-stranded
DNA1 is created

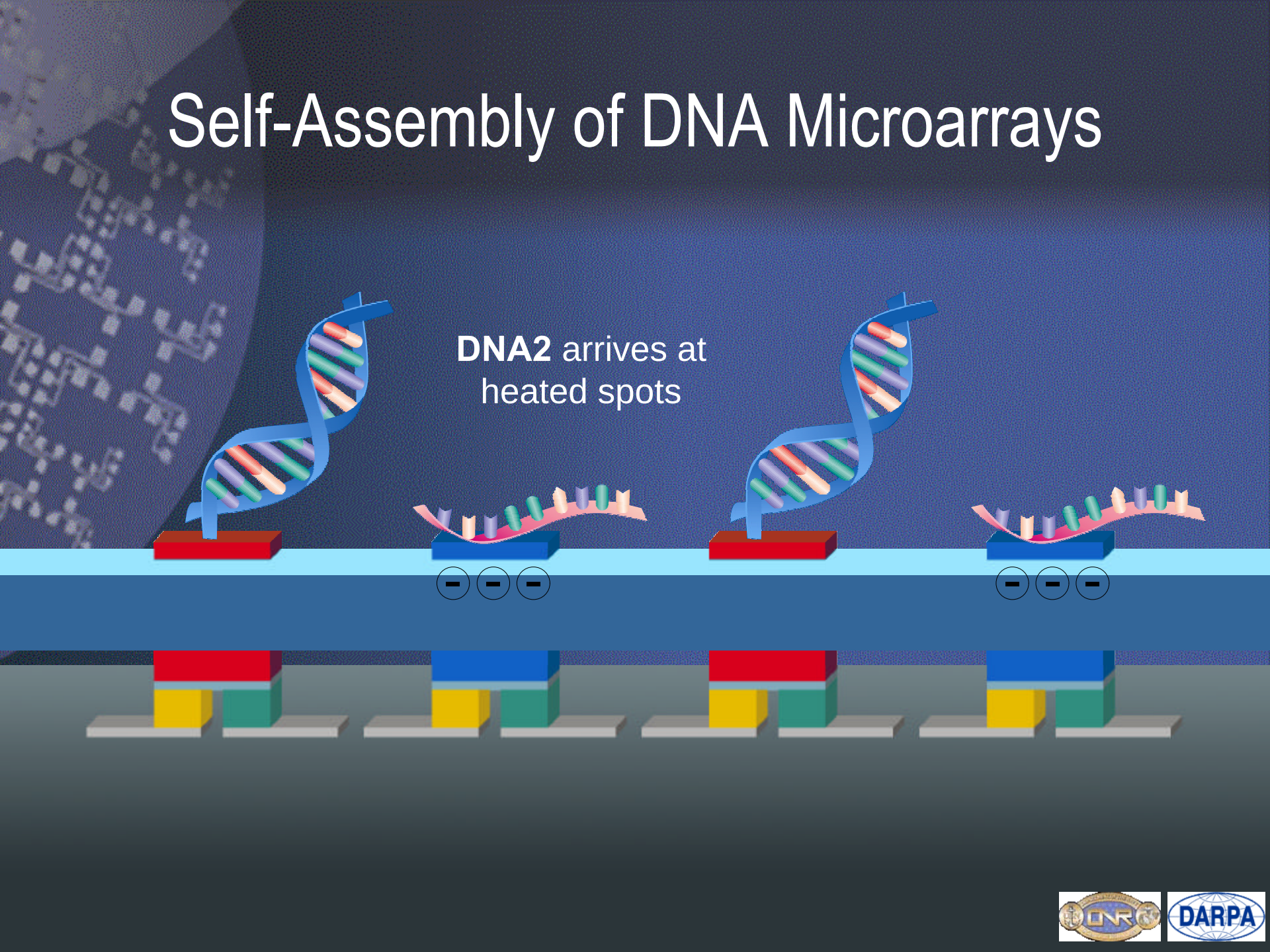


Self-Assembly of DNA Microarrays

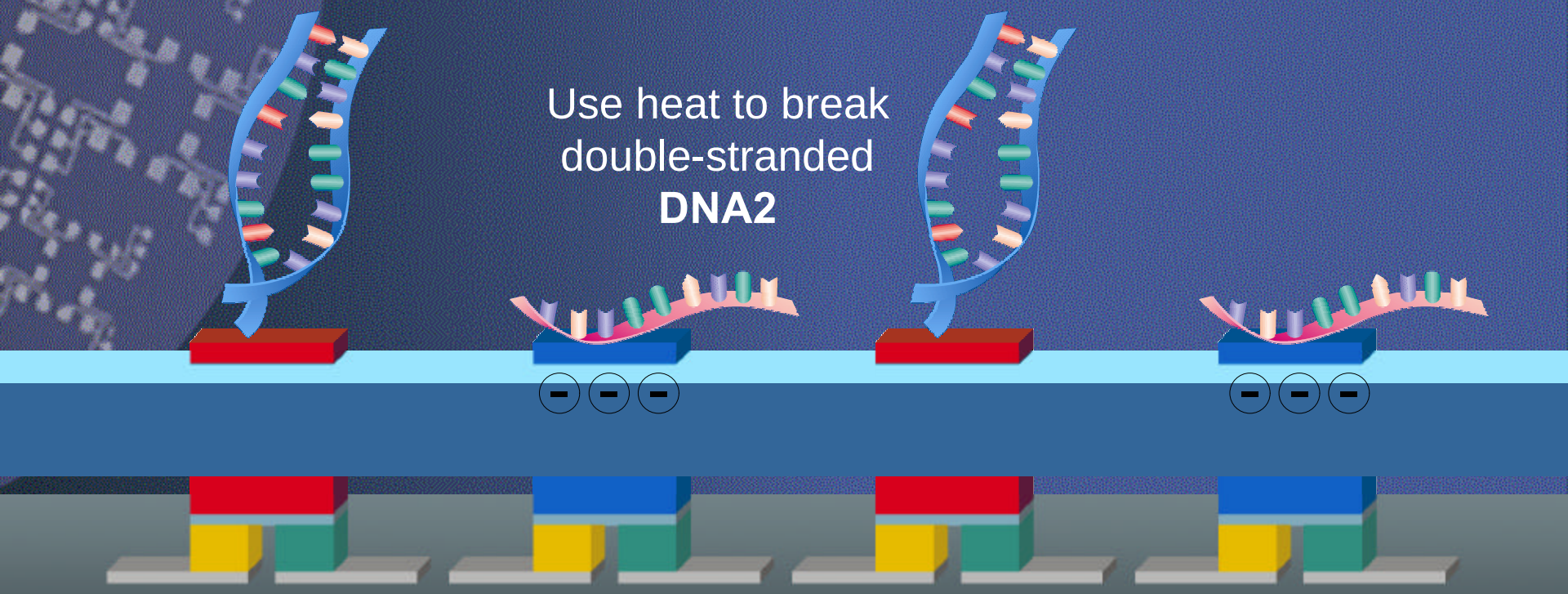
Use charge and cooling to trap single-stranded **DNA1**



Self-Assembly of DNA Microarrays

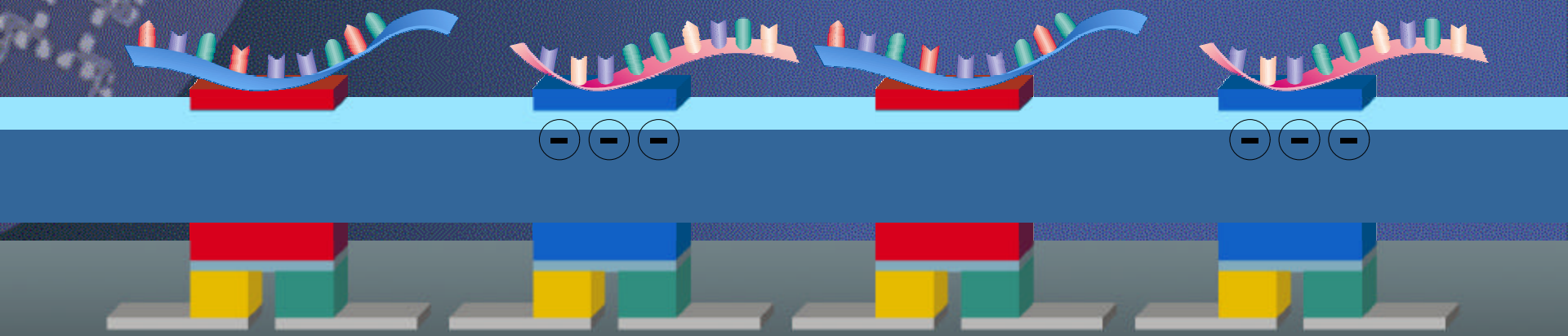


Self-Assembly of DNA Microarrays



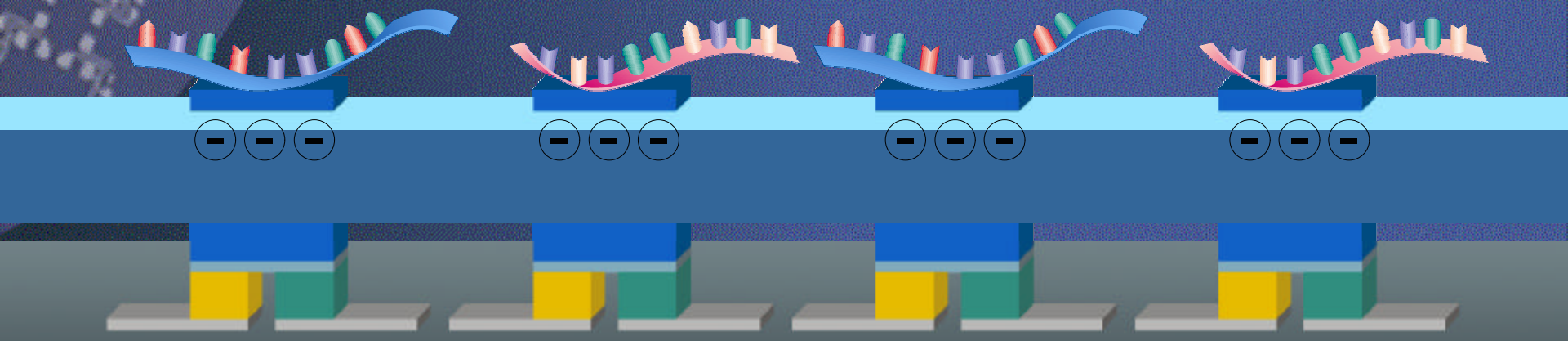
Self-Assembly of DNA Microarrays

A single-stranded
DNA2 is created



Self-Assembly of DNA Microarrays

Use charge and cooling to trap single-stranded **DNA2**

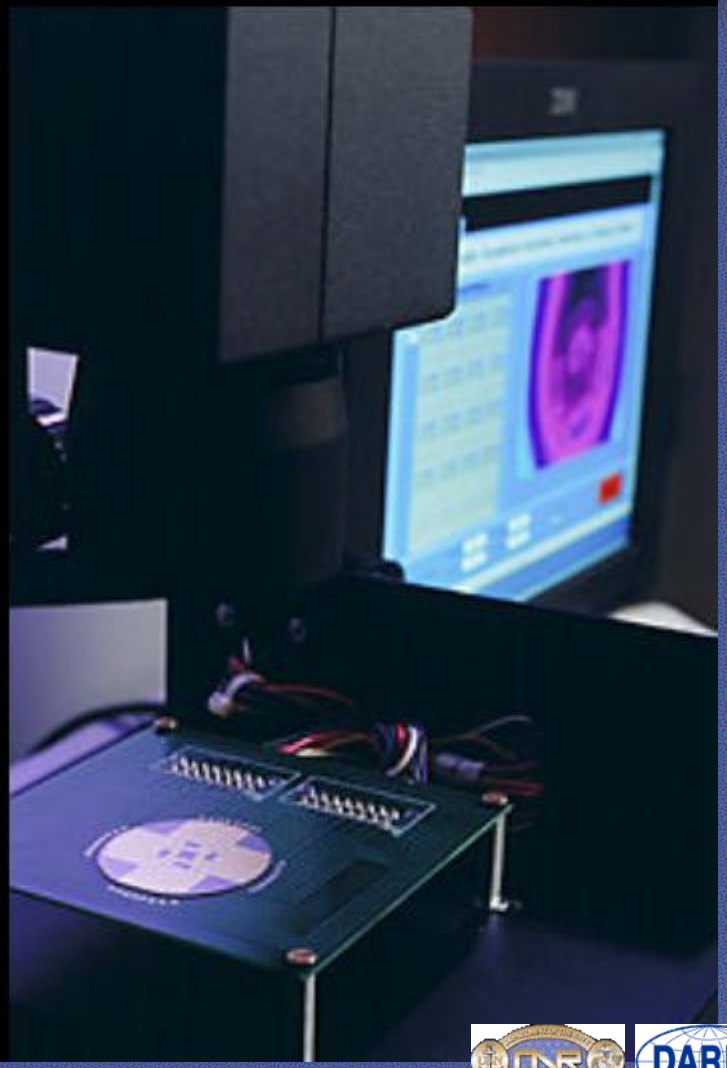


Self-Assembly of DNA Microarrays

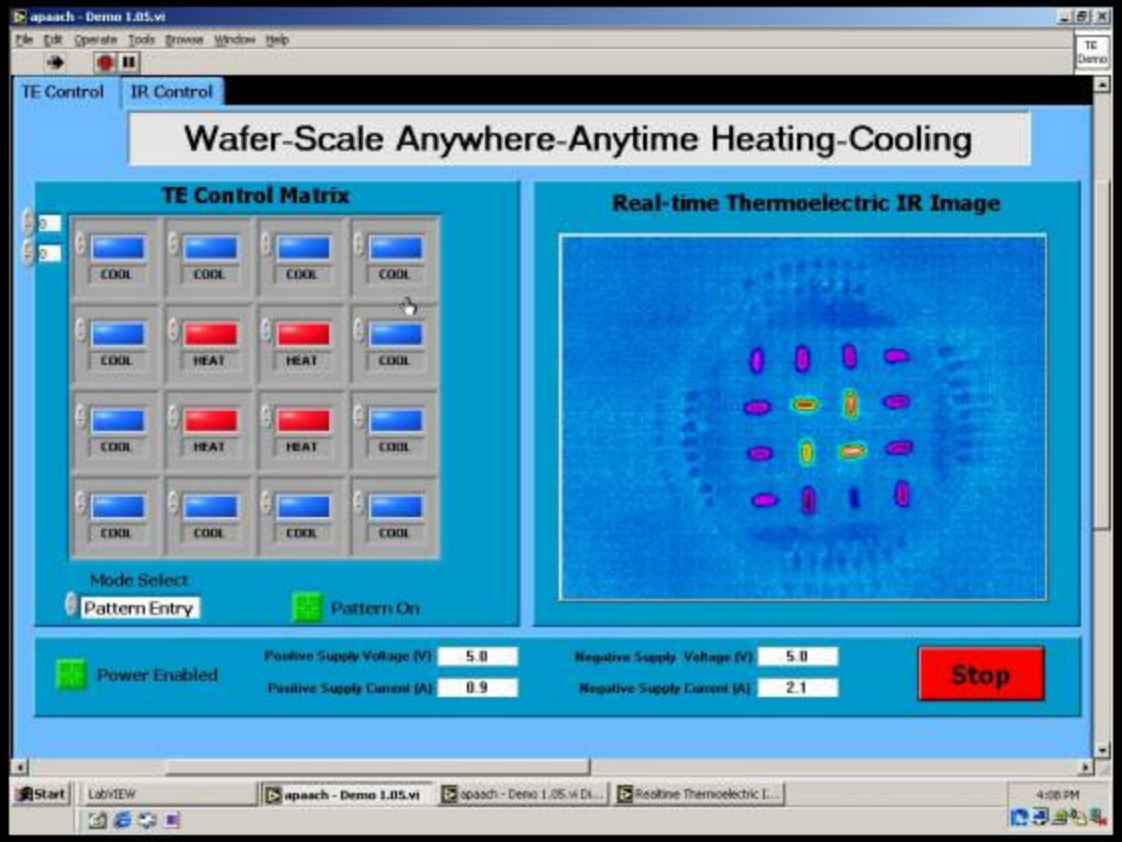
Self-assembled DNA-array ready
for experiments

RTI-Patent Pending

Wafer-Scale Spot-Temperature Control TE Wafer and Associated System Development

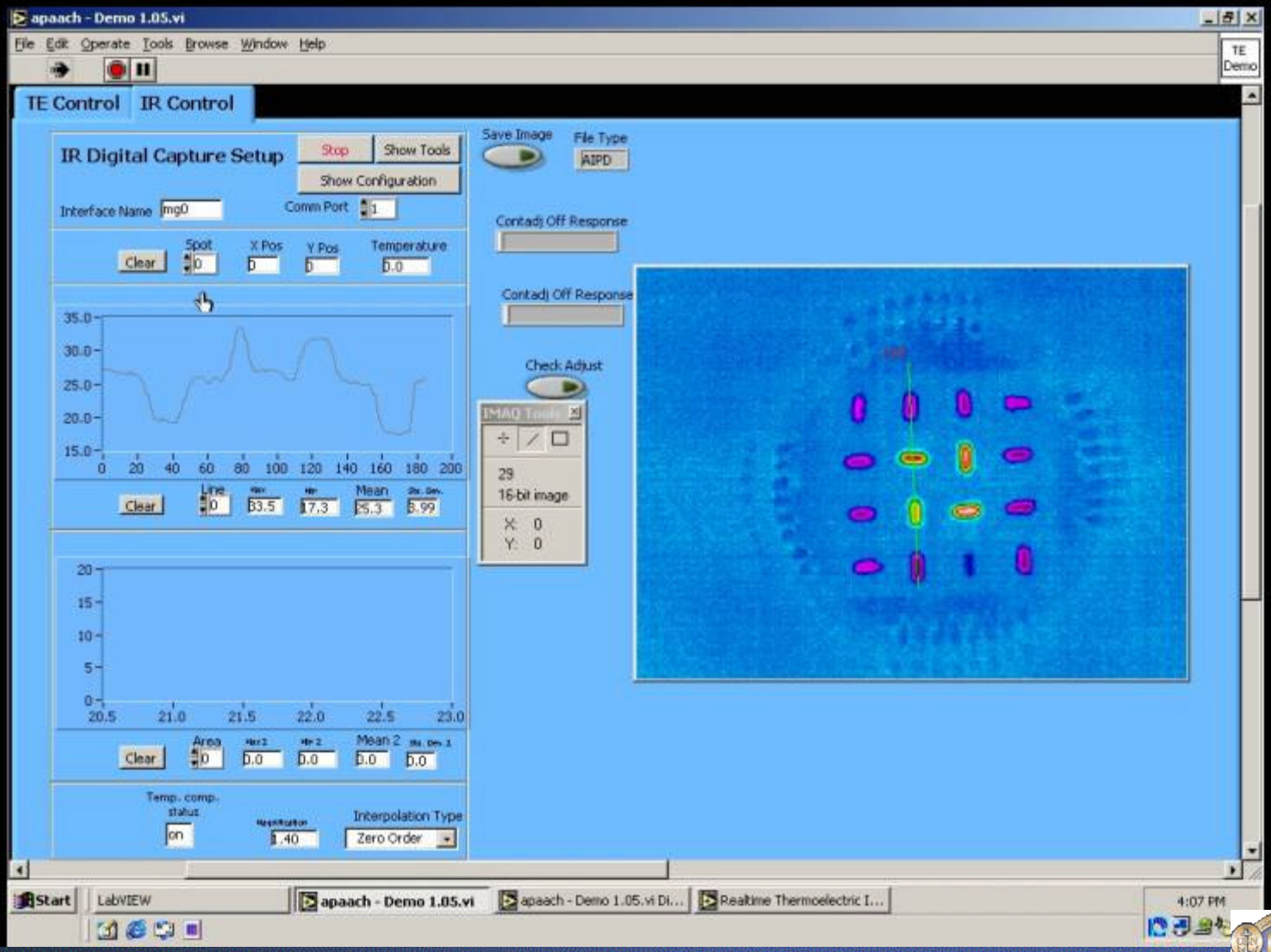


Wafer-Scale Spot-Temperature Control Using TE Devices (4 x 4 matrix using bulk devices)

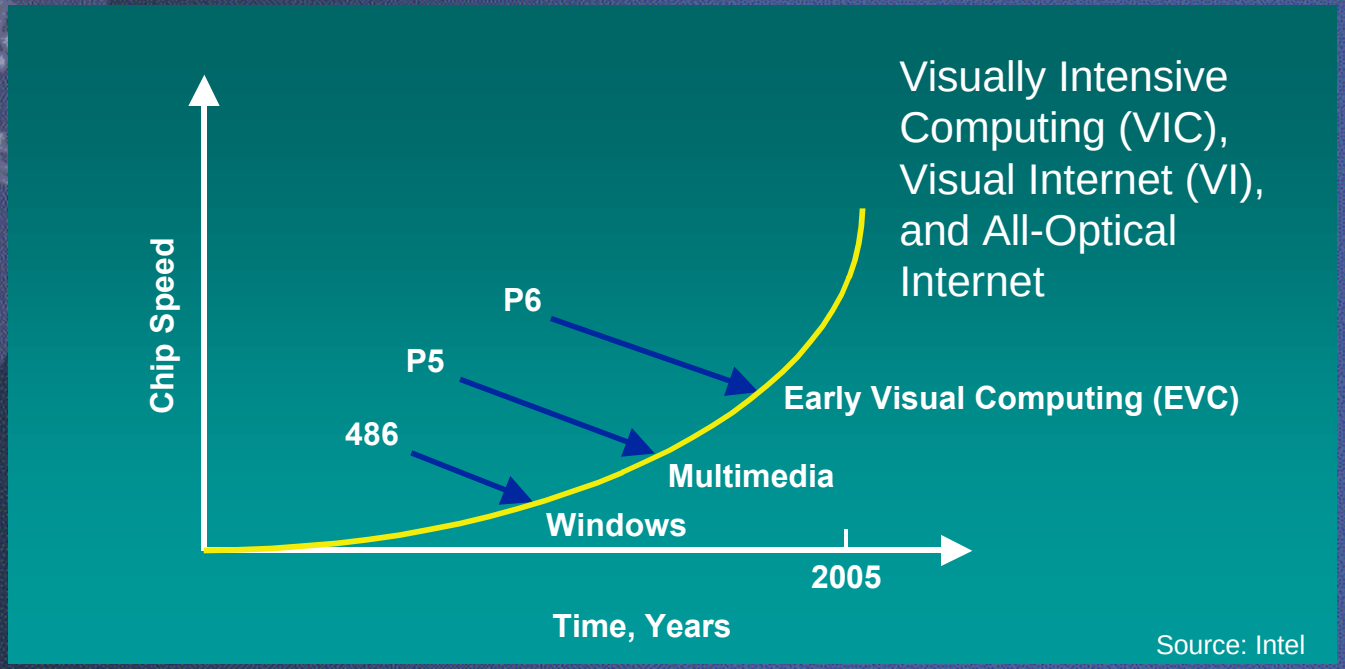


Working to control the individual temperature of each well of a typical 96-well plate for “Lab-on-a-chip” applications

Wafer-Scale Spot-Temperature Control Using TE Devices (4 x 4 matrix using bulk devices)



Chip Cooling for Complex Computation

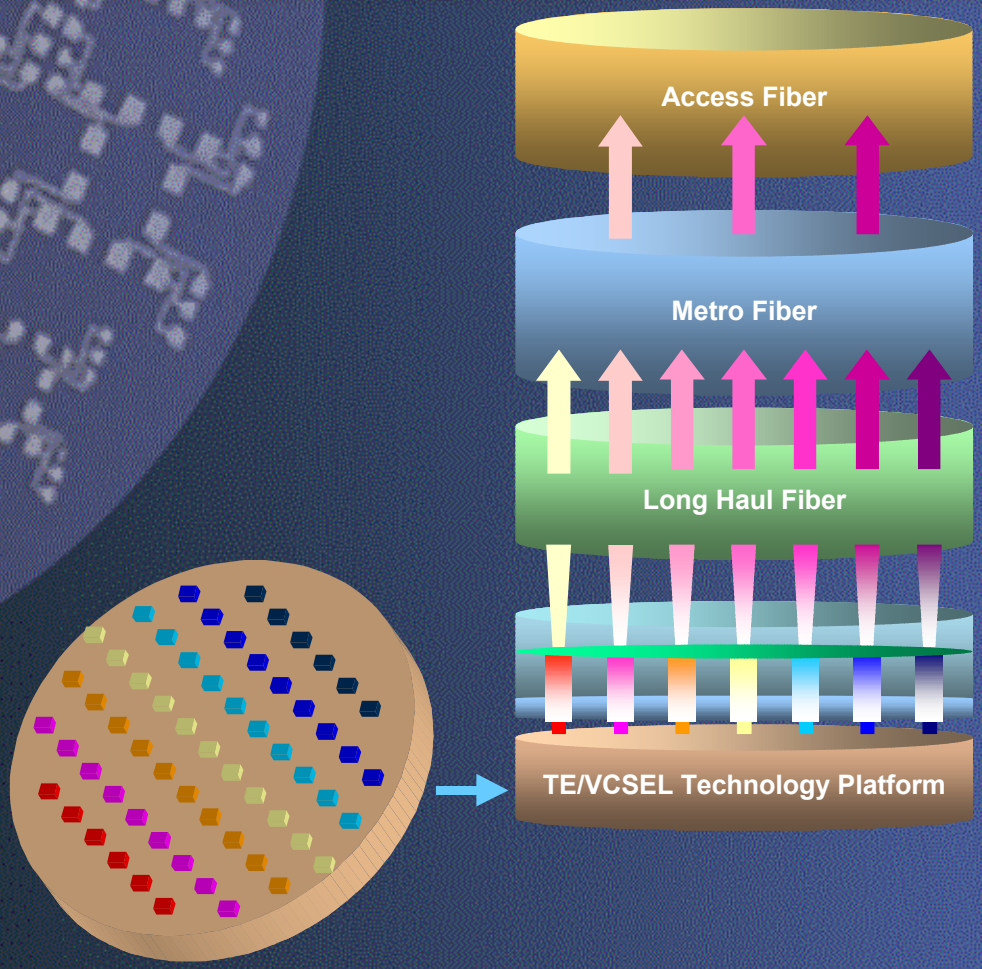


"If nothing changes, these chips will produce as much heat, for their proportional size, as a nuclear reactor. We have a huge problem to cool these devices."

— Pat Gelsinger, Intel CTO

February 5, 2001

Photonic Applications



Dynamic laser wavelength switching capability for payload transparency in large bandwidth transmission

Bulk versus Thin-film TE Power

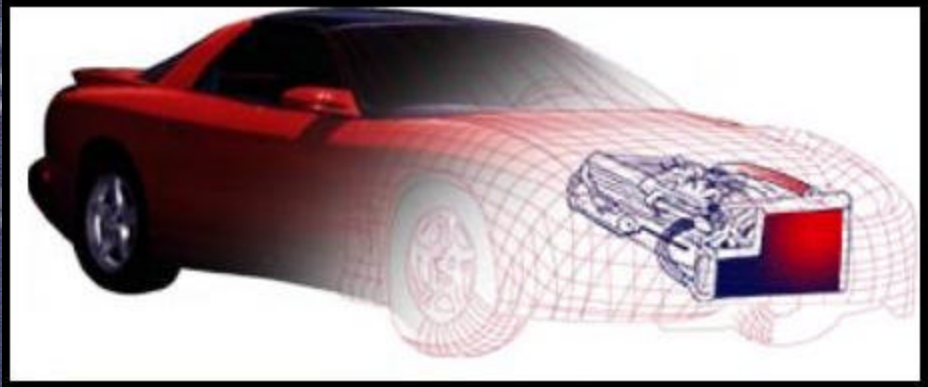
Performance Parameter	Bulk Technology (ZT ~ 1)	Thin-film Technology (ZT ~ 2.5)
Efficiency for a D**T of 150°C**	3 to 4%	10 to 11%
Specific Power excluding thermal management components	1 Watt/gram of TE module weight	570 Watts/gram of TE module weight
Cost	$5 per Watt as of today after 40 years of maturity	Long-term (three to five years) projected cost < $1 per Watt
Current Density Voltage levels from <u>each</u> module Power Density	< 200 mA/cm^2 5 to 10 Volts < 1 W/cm^2	> 50 Amp/cm^2 possible 48 Volts achievable with microelectronically interconnected circuits > 20 W/cm^2

Bulk versus Thin-film TE Cooling/Heating

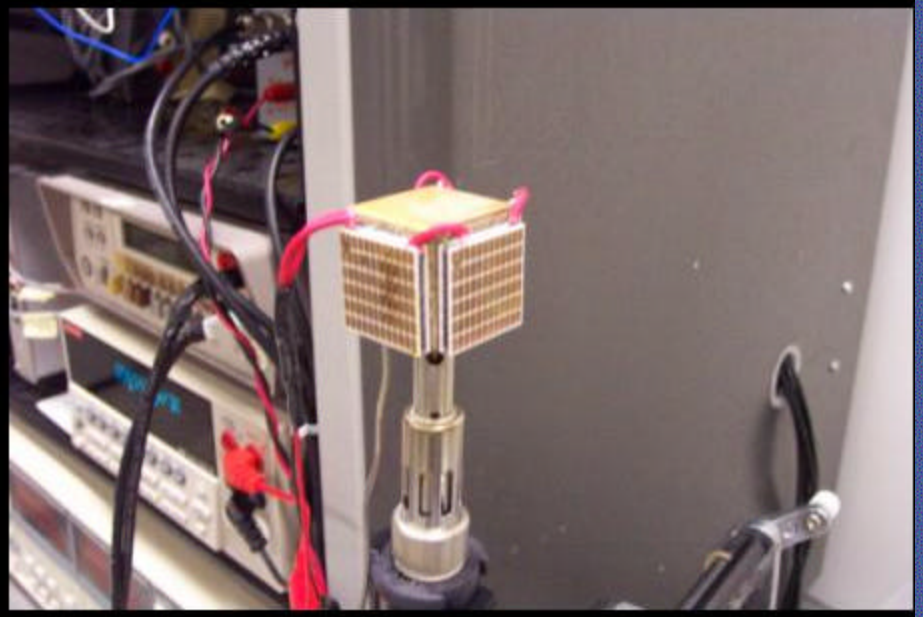
Performance Parameter	Bulk Technology (ZT ~ 1)	Thin-film Technology (ZT ~ 2.5)
COP (Heat Pumped/Electrical Power in) for a D**T of 25°C**	1	2 to 2.5
Wight of TE module for pumping 50 Watts (without thermal management components)	~ 22 grams	< 3 grams
Cost of heat pumping	$ 0.5 to 1 per Watt as of today after 40 years of maturity	Long-term (three to five years) projected cost < $ 0.5 per Watt
Cooling Power Density Potential Maximum Cooling per stage	< 1 W/cm^2 ~ 60°C	> 700 W/cm^2 ~ 120°C

Automotive Power

Waste-heat recovery for electrical power

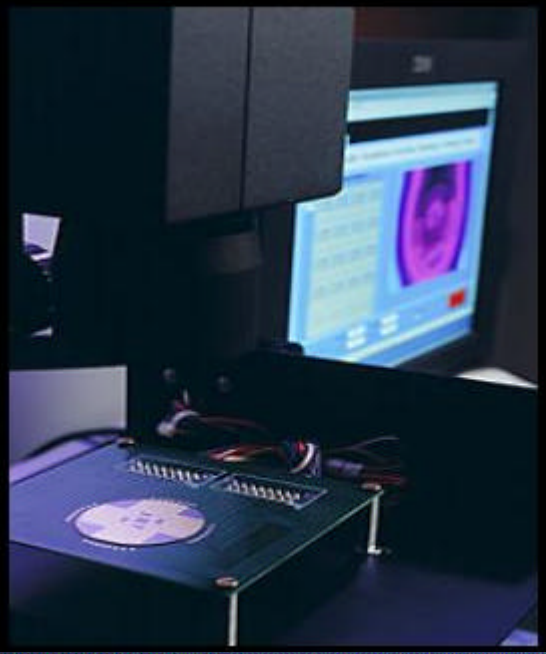


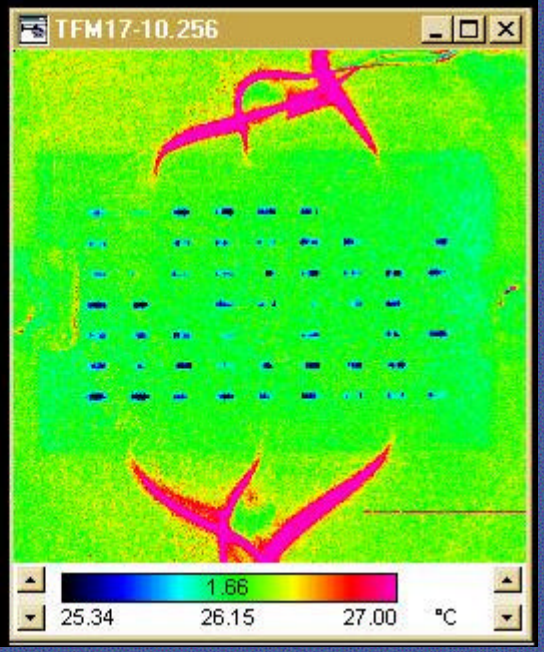
Portable power



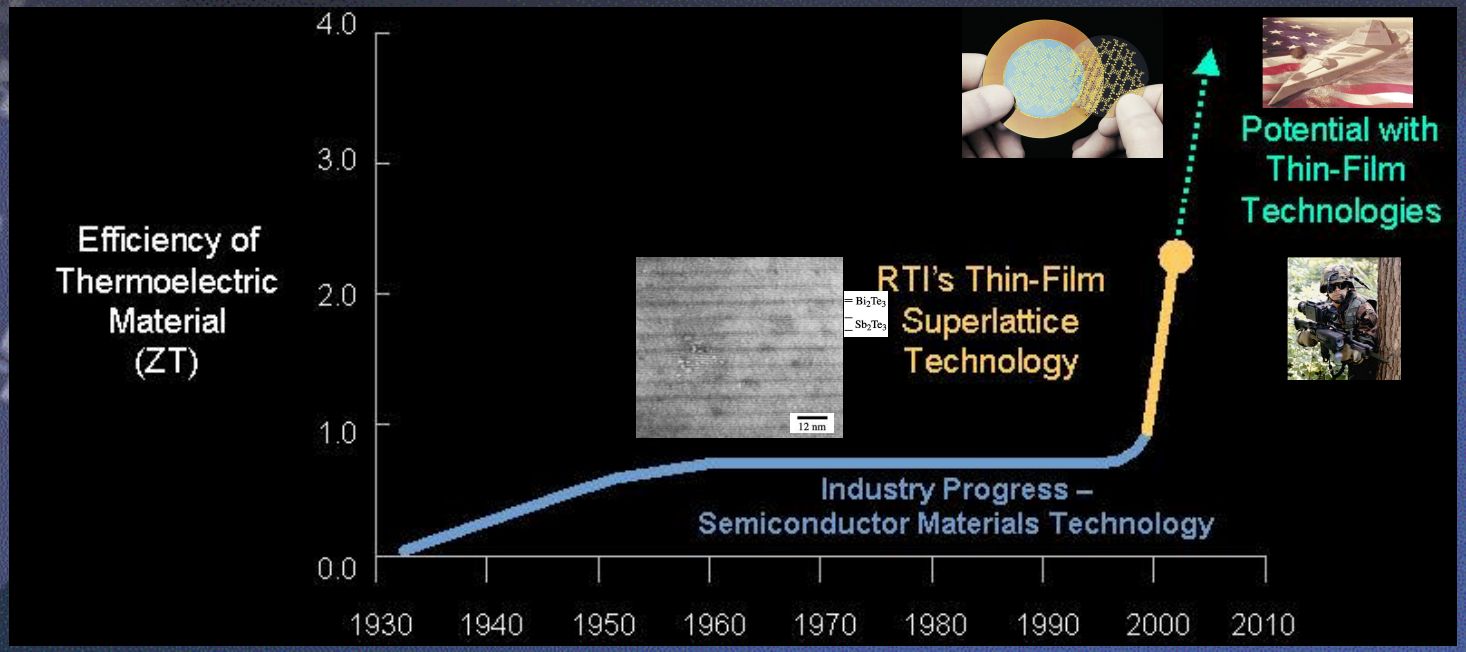
Thermal scavenging
for remote power

Automotive Computer-Based Spot Temperature Control for Electronics, Battery, etc.





Summary



- p-type Bi_2Te_3 /Sb_2Te_3 Superlattices : ZT of device ~2.4 and intrinsic ZT in the range of 2.92 at 400K
- n-type Bi_2Te_3 /$Bi_2Te_{3-x}Se_x$ Superlattices : ZT of device ~1.2 and intrinsic ZT in the range of 1.4 at 300K
- Thin-film devices can lead to high power density, fast response, wafer-scale integration for both cooling and power conversion