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ELASTIC AND ACTIVATED THERMOELECTRIC TRANSPORT AT THE BAND EDGES OF DISORDERED NANOWIRES

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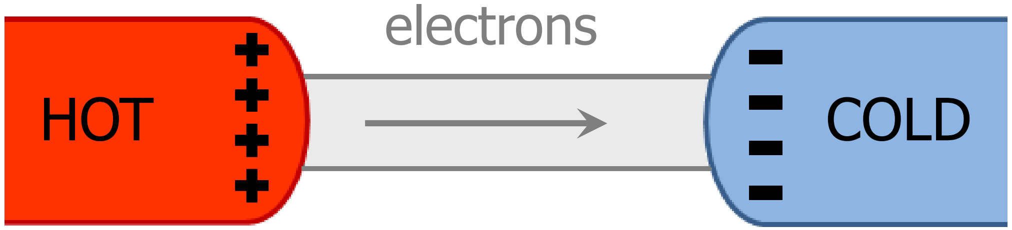
Service de Physique de l'Etat Condensé

References

- **Riccardo Bosisio, PhD Thesis, Univ Paris 6 (sept 2014)**
- **Riccardo Bosisio, Geneviève Fleury** and JLP, New Journal of Physics 16 (2014) 035004
- **Riccardo Bosisio, Cosimo Gorini, Geneviève Fleury** and JLP, New Journal of Physics 16 (2014) 095005
- **Riccardo Bosisio, Cosimo Gorini, Geneviève Fleury** and JLP, arXiv:1407.7020

Luchon, March 2015

Thermoelectricity : Rules of the game



Thermopower S (or Seebeck coeff.):

$$S = -\left(\frac{\Delta V}{\Delta T}\right)_{J^e=0}$$

Maximize the efficiency i.e. the figure of merit :

$$ZT = \frac{GS^2}{K^e + K^{ph}} T$$

… keeping a reasonable electrical output power (power factor) :

$$Q = GS^2$$

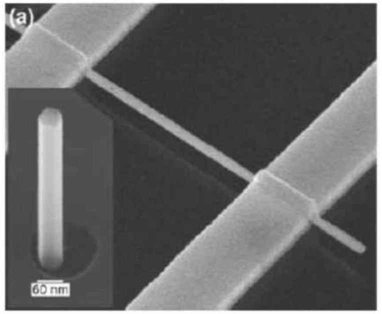
Why semiconductor nanowires?

"… a newly emerging field of low-dimensional thermoelectricity, enabled by materials nanoscience and nanotechnology"

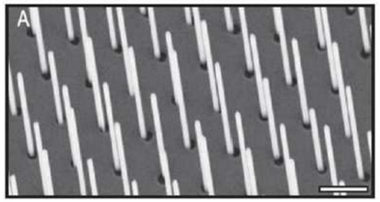
Dresselhaus et al: Adv. Mater. 2007

"… fundamental scientific challenges could be overcome by deeper understanding of charge and heat transport"

Majumdar: Science 2004



SC nanowires



Reduced thermal conductance

Phonon vs electrons mean free path, geometrical designs *(Hochbaum 2008, Heron 2010)*

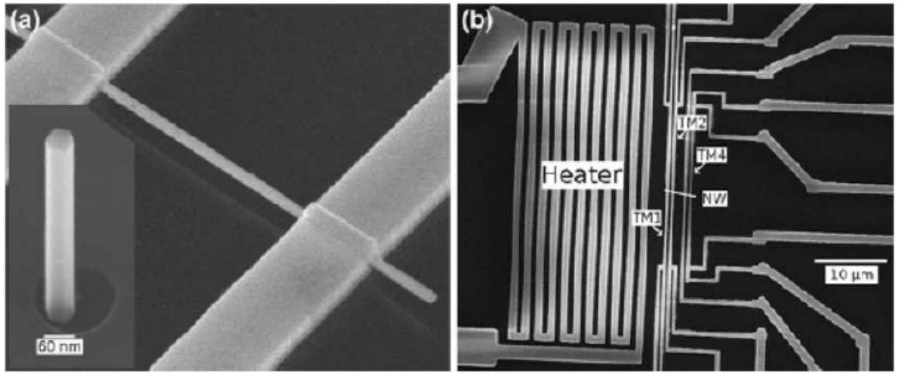
Enhanced thermopower

Field effect transistors *(Brovman 2013, Roddaro 2013 & many others)*

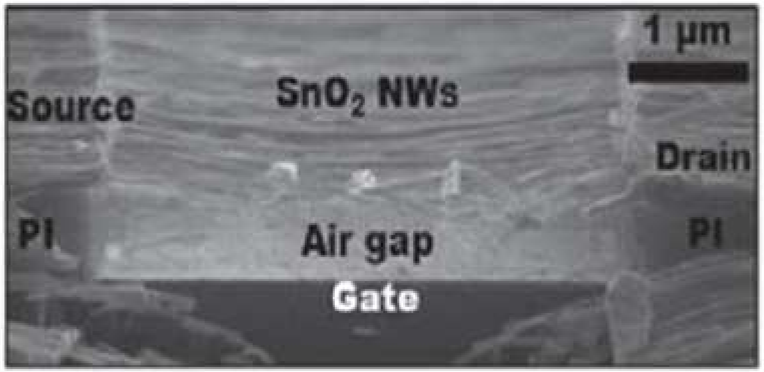
Scalable output power

Arrays of parallel NWs *(Pregl 2013, Stranz 2013)*

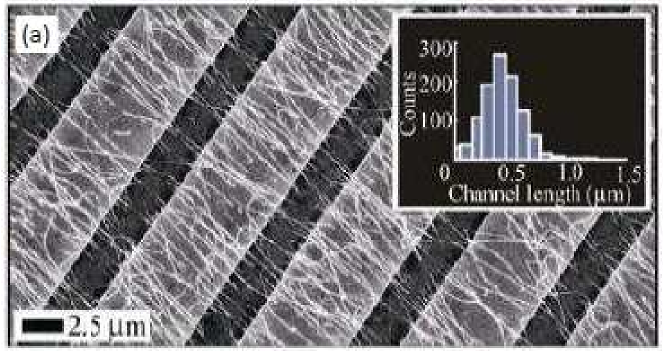
Some experimental realizations



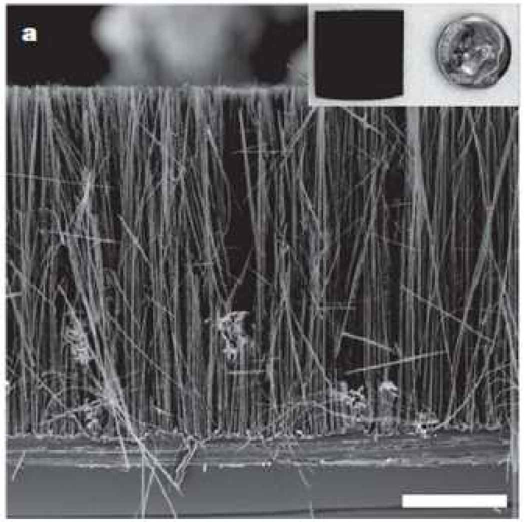
Karg *et al.* (IBM Zurich), 2013



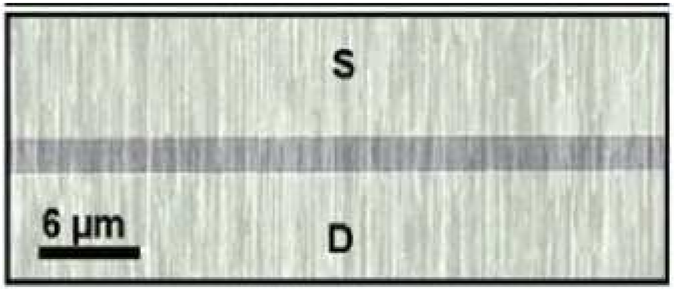
Shin *et al.* (Seoul), 2011



Pregl *et al.* (TU Dresden), 2013



Hochbaum *et al.* (Berkeley CA), 2008



Fan *et al.* (Berkeley CA), 2008

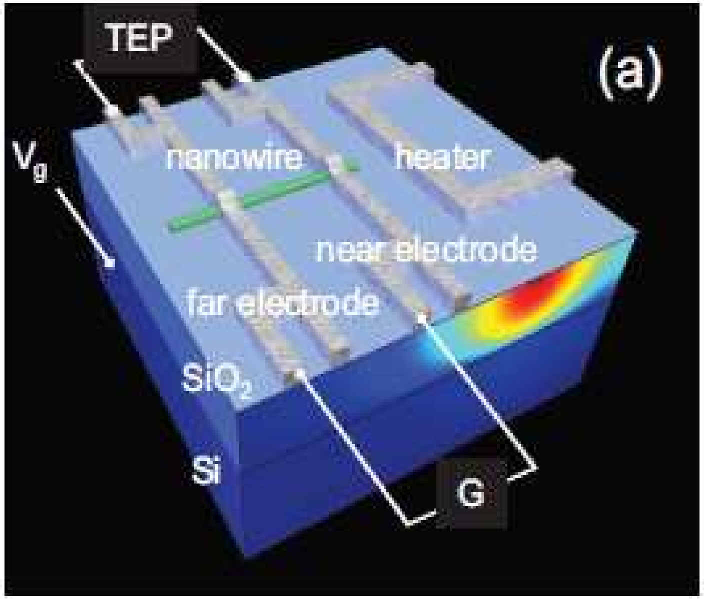
Many experimental works and a few theoretical works

Outline

Nanowire in the Field Effect Transistor Device Configuration
described with a 1D Anderson Model
(tight binding 1d lattice with constant hopping and random site potentials)

- ❑ 1. Thermopower of single NW: **low T elastic (tunnel)** regime
- ❑ 2. Thermopower of single NW: **Intermediate T inelastic phonon-assisted** regime (Mott variable range hopping)
- ❑ 3. Large **Arrays of Parallel NWs**: Applications for
- Field control of the phonons at sub-micron scales (heat management)
- Energy harvesting (transforming the waste heat into useful electrical power)
- Hot spot cooling (important for microelectronics)

Field-Effect Transistors (FET)



Setup used by P. Kim (Columbia) (2013)

- Single (or array of) doped **nanowire(s)** in the FET configuration
- **Substrate**: Electrically and thermally insulating
- **Gate**: «back» or «top»
- **Heater**: for thermoelectric measurements

"Seebeck" configuration: thermal bias

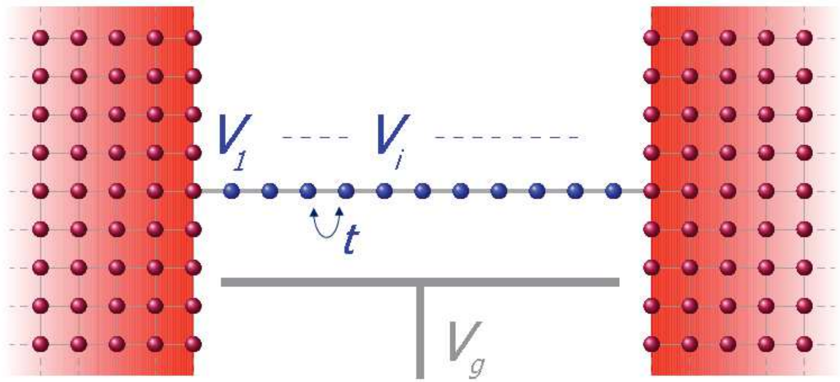
"Peltier" configuration: voltage bias

⟹ $\Pi = ST$: equivalent within linear response if time-reversal symmetry preserved (Kelvin-Onsager relation)

Goal: Control of the thermopower with the back gate

1D Anderson Model

Prototypical model of localized system

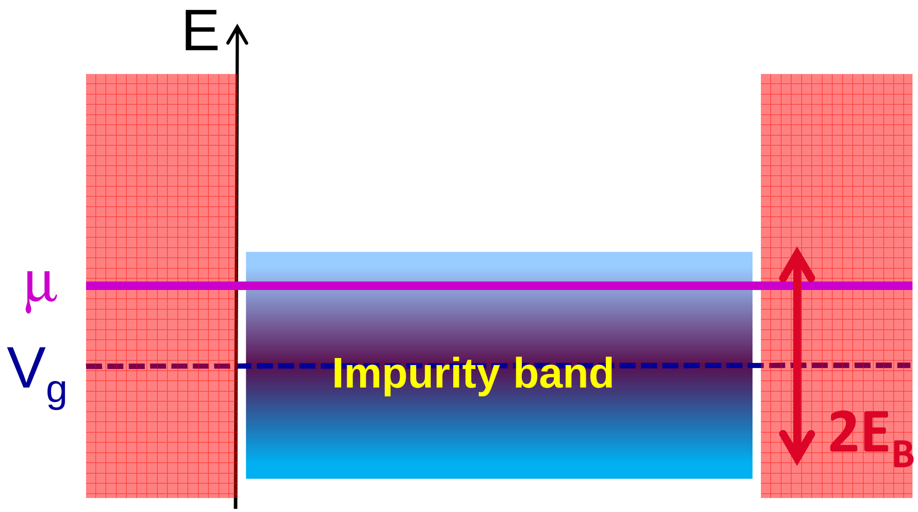


Gate Voltage

- 1D electronic lattice with on-site (uniform) disorder $V_i \in$ (-W/2, W/2)

- Tight-binding Hamiltonian

$$\mathcal{H} = -t \sum_{i=1}^{N-1} \left(c_i^\dagger c_{i+1} + \text{h.c.} \right) + \sum_{i=1}^{N} (V_i + V_g) c_i^\dagger c_i$$



- All electrons are localized with localization length ξ (E)

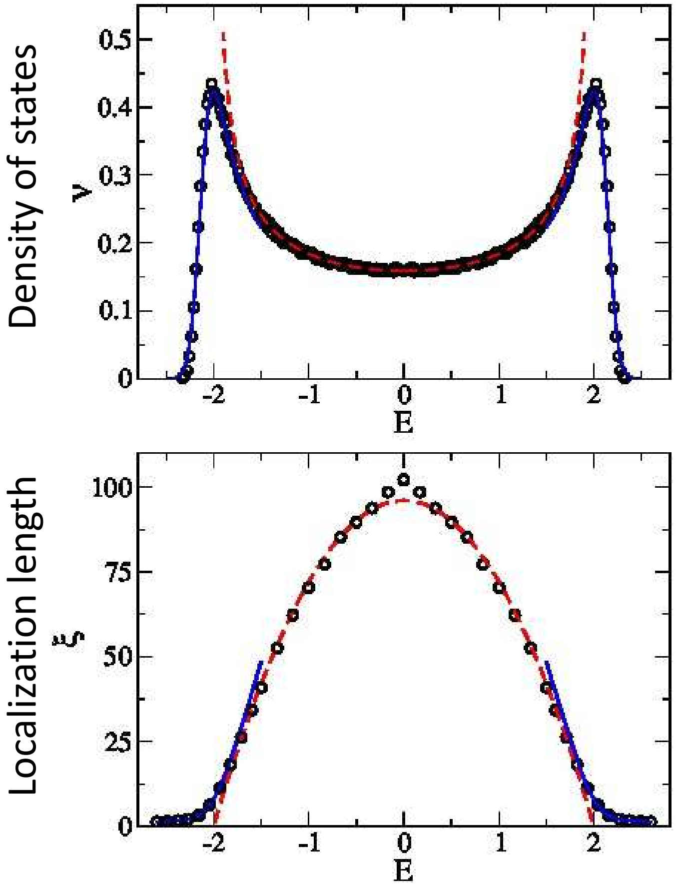
- States distributed within an impurity band of width $2E_B$≈4t+W

- Behavior of the typical thermopower when the gate voltage V_g is varied

1D density of states ν and localization length ξ

$V_G = 0$ Analytical expressions derived in the **weak disorder limit**

W=t : band edge at
~ 2t+W/2 ~ 2.5t



"Bulk" formulas:

$$\nu_b(E) = \frac{1}{2\pi t\sqrt{1-(E/2t)^2}}$$

$$\xi_b(E) = \frac{24}{W^2}\left(4t^2 - E^2\right)$$

"Edge" formulas:

$$\nu_e(E) = \sqrt{\frac{2}{\pi}}\left(\frac{12}{tW^2}\right)^{1/3}\frac{\mathcal{I}_1(X)}{[\mathcal{I}_{-1}(X)]^2}$$

$$\xi_e(E) = 2\left(\frac{12t^2}{W^2}\right)^{1/3}\frac{\mathcal{I}_{-1}(X)}{\mathcal{I}_1(X)}$$

$$\mathcal{I}_n(X) = \int_0^{\infty} y^{n/2}\, e^{-\frac{1}{6}y^3+2Xy}\, dy$$

$$X = (|E| - 2t)t^{1/3}(12/W^2)^{2/3}$$

B. Derrida & E. Gardner, *J. Physique* 45, 1283 (1984)

1. Elastic regime: Thermopower

Theory: Transport mechanism: elastic (coherent) tunnelling

Localized regime: τ decays exponentially with length

$$[\ln \mathcal{T}]_0(E) = -\frac{2N}{\xi(E)}$$

Typical τ depends on the energy via ξ(E) (**localization length**)

Low Temperatures + Linear Response → Mott formula:

$$S \approx \frac{\pi^2}{3}\frac{k_B}{e} k_B T \left.\frac{\mathrm{d}\ln \mathcal{T}}{\mathrm{d}E}\right|_{\mu}$$

Numerics: Recursive Green Function calculation of S

Elastic Regime: Typical Thermopower

Bulk:

$$S_0^b = N \frac{(\mu - V_g)\, W^2}{96 t^3 [1 - ((\mu - V_g)/2t)^2]^2}$$

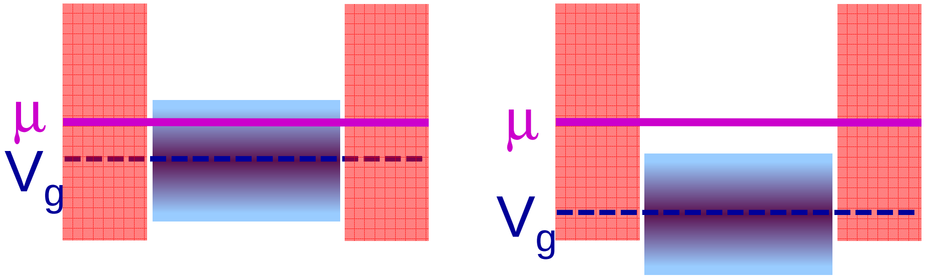
Edge:

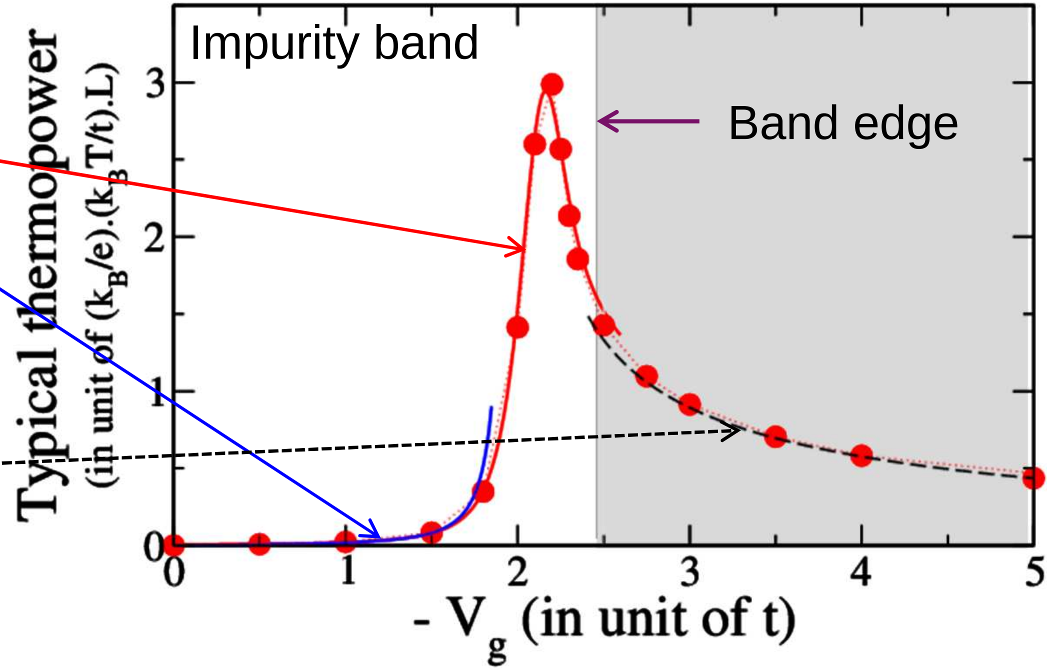
$$S_0^e = 2N \left(\frac{12t^2}{W^2}\right)^{1/3} \left\{ \frac{\mathcal{I}_3(X)}{\mathcal{I}_{-1}(X)} - \left[\frac{\mathcal{I}_1(X)}{\mathcal{I}_{-1}(X)}\right]^2 \right\}$$

$$X = (|\mu - V_g| - 2t)\, t^{1/3} (12/W^2)^{2/3}$$

Tunnel Barrier:

$$\frac{S_0^{TB}}{N} \underset{N\to\infty}{\approx} -\frac{1}{N}\frac{2t}{\Gamma(\mu)}\frac{d\Gamma}{dE}\Big|_{\mu} \mp \frac{1}{\sqrt{\left(\frac{\mu - V_g}{2t}\right)^2 - 1}}$$

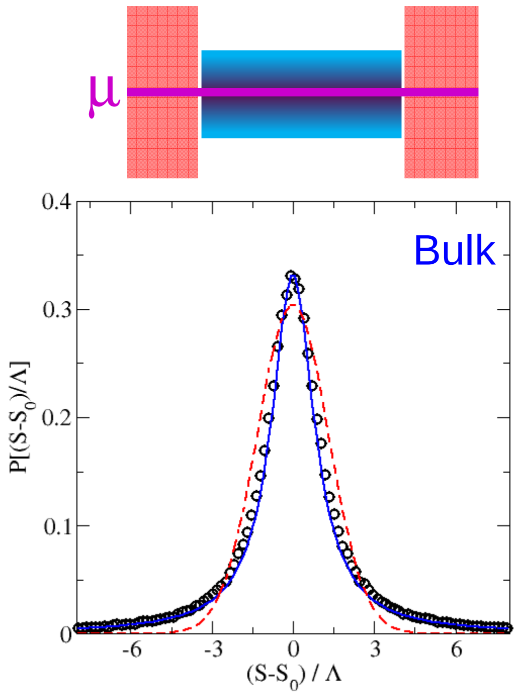




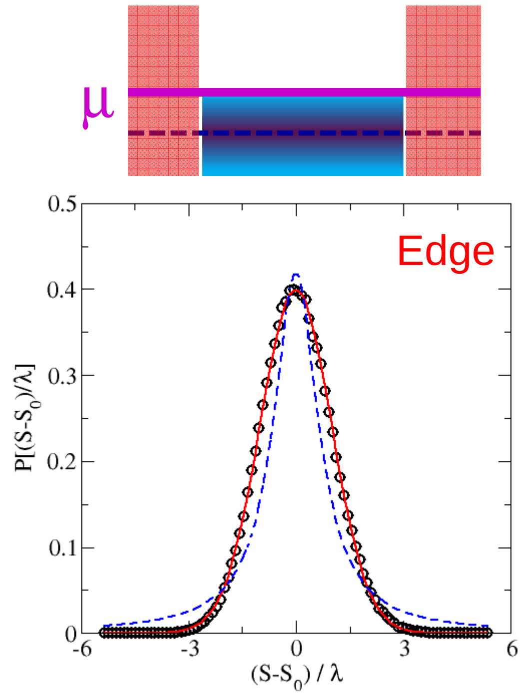
Large increase of the (typical) thermopower near the band edge,
perfectly well described analytically

R. Bosisio, G. Fleury, & J.-L. Pichard, *New J. Phys*. 16:035004 (2014)

Elastic Regime: Fluctuations



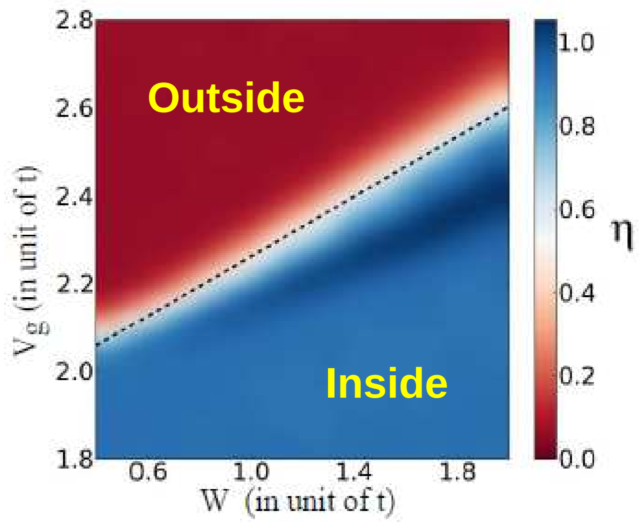
Lorentzian



Gaussian

Transition:

$$\eta = \frac{\int d\mathcal{S}|P(\mathcal{S}) - P_G(\mathcal{S})|}{\int d\mathcal{S}|P_L(\mathcal{S}) - P_G(\mathcal{S})|}$$



- Lorentzian : $P(\mathcal{S}) = \frac{1}{\pi}\frac{\Lambda}{\Lambda^2 + (\mathcal{S} - \mathcal{S}_0)^2}$ with $\Lambda = \frac{2\pi t}{\Delta_F}$ (Δ_F mean level spacing)
- Gaussian : $P(\mathcal{S}) = \frac{1}{\sqrt{2\pi}\lambda}\exp\left[-\frac{(\mathcal{S} - \mathcal{S}_0)^2}{2\lambda^2}\right]$ with $\lambda \approx 0.6\frac{Wt\sqrt{N}}{(\mu - V_g)^2 - (2t + W/4)^2}$

S. A. van Langen, P. G. Silvestrov, & C.W. J. Beenakker, *Superlattices Microstruct*. 23, 691 (1998).

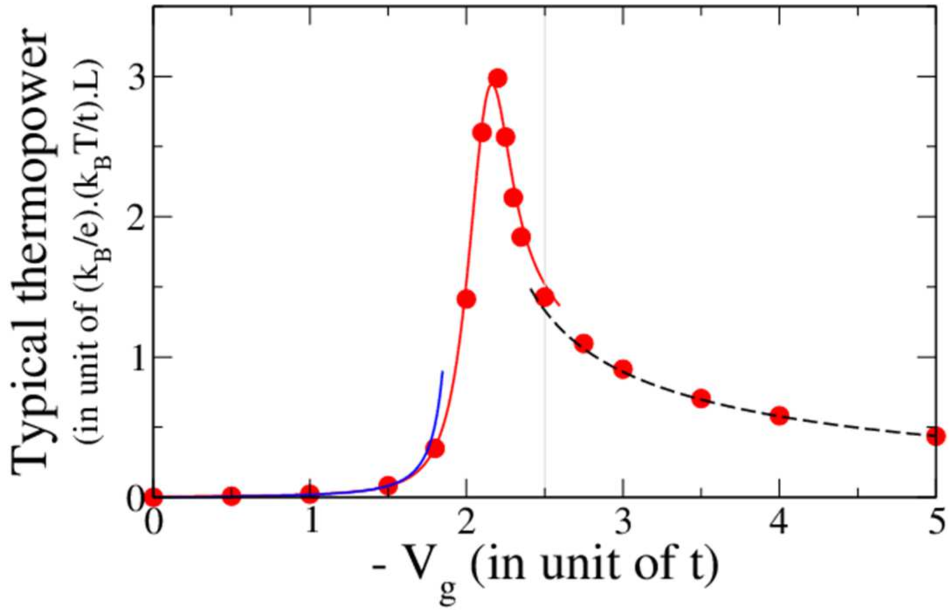
Elastic regime: Summary

Enhancement of the thermopower at the band edges (role of $\xi(E)$)

Analytical description of the results

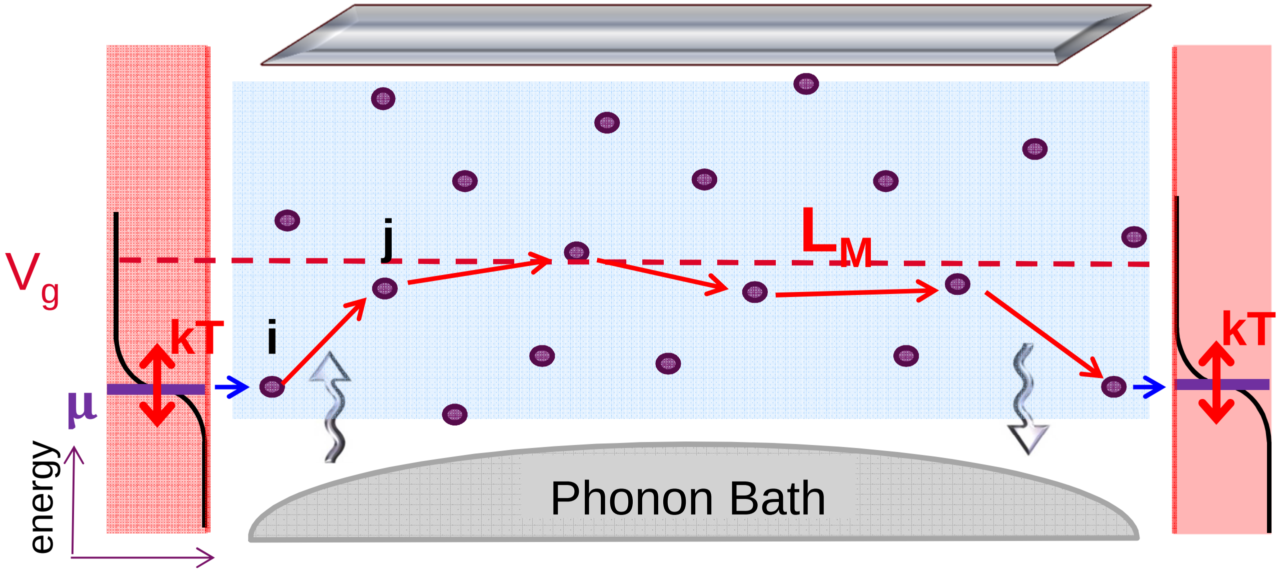
Sommerfeld Expansion (low T)
Wiedemann-Franz law → Low S

Very low power factor $Q = GS^2$ because of the exponential reduction of G at the band edges



Interest: Ultra-low T : Peltier cooling?

OR → toward higher temperatures!



Mott → competition between tunneling and activated processes

$$G_{ij} \sim e^{-2|x_i - x_j|/\xi}\, e^{-(|E_i-\mu|+|E_j-\mu|+|E_i-E_j|)/2k_BT}$$

Variable Range Hopping: phonon-assisted transport → *sequence of hops of variable size*

Optimal hop size: *Mott hopping length*

$$L_M \simeq \left(\frac{\xi}{2\nu T}\right)^{1/2}$$

or *Mott hopping energy*

$$\Delta = \left(\frac{2T}{\xi\nu}\right)^{1/2}$$

$$\rightarrow G \sim e^{-\frac{2L_M}{\xi}} = e^{-\frac{\Delta}{k_BT}}$$

Transport Mechanisms

$$L_M \simeq \left(\frac{\xi}{2\nu T}\right)^{1/2}$$

T ↓

Low T: $L \ll L_M$ → elastic coherent transport

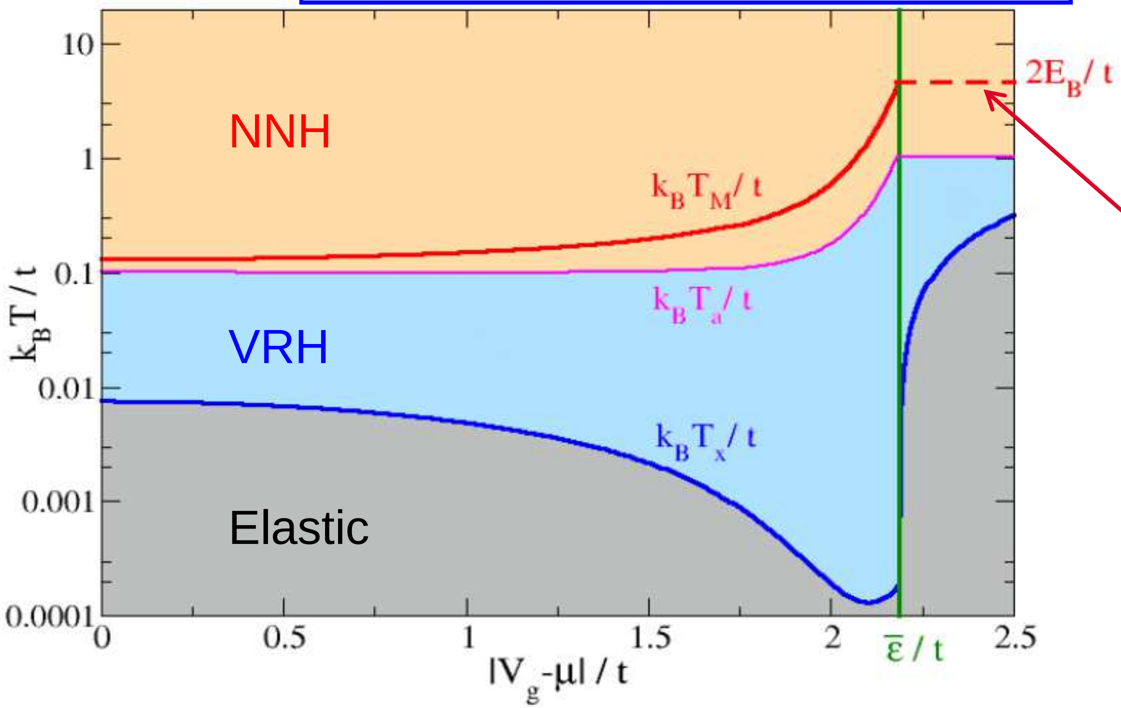
Increasing T: $L_M \sim L$ → onset of inelastic processes (VRH) → $T_x \simeq \frac{\xi}{2\nu L^2}$

Increasing T: $L_M \sim \xi$ → simple activated transport (NNH) → $T_M \simeq \frac{2}{\xi\nu}$

Mott's Hopping Energy:
finite range of states contributing to transport

$$\Delta = k_B\sqrt{TT_M} \propto \sqrt{\frac{T}{\xi\nu}} \gg k_B T$$

Relevant energy scale for activated transport



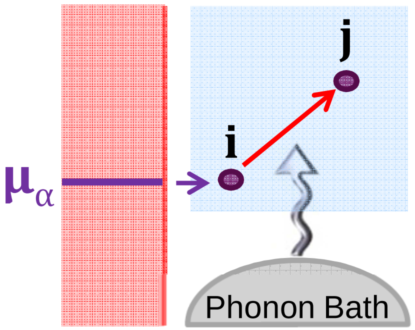
Cut-off required by $\Delta(T_M) = k_BT_M \leq 2E_B$

(T_a : onset of simple activation in 1D (Kurkijärvi 1973, Raikh & Ruzin 1989))

Inelastic (Phonon-Assisted) Regime: method

Essential ingredients to build & solve the Random Resistor Network

1. **Transition rates (Fermi golden rule)**



Between lead and localized states **[Elastic tunneling rates]**

$$\Gamma_{i\alpha} = \gamma_{i\alpha} f_i \, [1 - f_\alpha(E_i)] \quad \alpha = \text{L,R}$$

$$\gamma_{i\alpha} \simeq \gamma_e \exp(-2x_{i\alpha}/\xi_i)$$

Between localized states **[Inelastic hopping rates]**

$$\Gamma_{ij} = \gamma_{ij} f_i (1 - f_j) \, [N_{ij} + \theta(E_i - E_j)]$$

$$\gamma_{ij} \simeq \gamma_{ep} \exp(-2x_{ij}/\xi)$$

← ξ energy dependence usually neglected!

$$\rightarrow \quad \gamma_{ij} \propto |\langle\psi_i|\psi_j\rangle|^2 \simeq \gamma_{ep} \left| \frac{(1/\xi_j)\exp(-x_{ij}/\xi_i) - (1/\xi_i)\exp(-x_{ij}/\xi_j)}{(1/\xi_i - 1/\xi_j)} \right|^2$$

J-H. Jiang, O. Entin-Wohlman, and **Y. Imry**, *Phys. Rev. B* 87:205420 (2014)

Inelastic (Phonon-Assisted) Regime: method

2. Fermi distributions at equilibrium (no bias)

$$f_i^0 = \left(e^{(E_i-\mu)/kT}+1\right)^{-1} \qquad f_\alpha = f_\alpha^0(E_i) = \left(e^{(E_i-\mu)/kT}+1\right)^{-1}$$

3. Occupation probabilities out of equilibrium

$$f_i = f_i^0 + \delta f_i \qquad f_\alpha = f_\alpha^0 + \delta f_\alpha$$

↑ ↑

4. Currents

$$I_{ij} = e\left(\Gamma_{ij} - \Gamma_{ji}\right) \qquad I_{i\alpha} = e\left(\Gamma_{i\alpha} - \Gamma_{\alpha i}\right)$$

5. Current conservation at every node i (Kirchoff)
N coupled equations in N variables

$$\left(\sum_{j\neq i} I_{ij}\right) + I_{iL} + I_{iR} = 0 \quad \rightarrow \quad \delta f_i$$

J-H. Jiang, O. Entin-Wohlman, and Y. Imry., *Phys. Rev. B* 87:205420 (2014)

Miller – Abrahams Resistor Network

6. Total particle/heat currents

Summing all terms flowing out from L(R) terminal

$$J_L^e = -\sum_i I_{iL} = \sum_i I_{iR},$$

$$J_{L(R)}^Q = \sum_i \left(\frac{E_i - \mu_{L(R)}}{e}\right) I_{L(R)i}.$$

7. Transport coefficients

$$G = \frac{J_L^e}{\delta\mu/e}$$ **Conductance**

$$\Pi = \frac{J_L^Q}{J_L^e}$$ **Peltier coefficient**

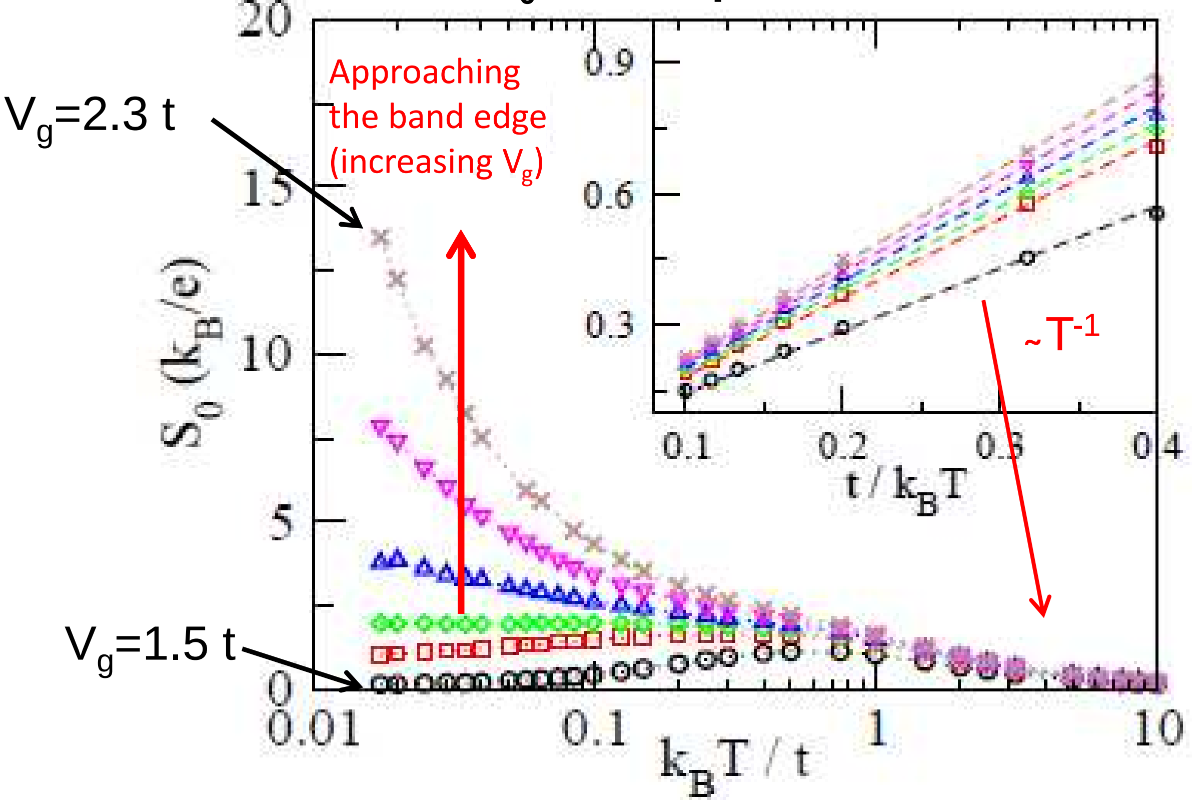
$$S = \frac{\Pi}{T}$$ **Thermopower**

(Having assumed Peltier configuration: T constant everywhere)

J-H. Jiang, O. Entin-Wohlman, and Y. Imry., *Phys. Rev. B* 87:205420 (2014)

Inelastic Regime: Typical thermopower

S_0 vs Temperature



- Thermopower enhancement when the band edges are approached
- Rich behaviour of the T-dependence of the thermopower, "reflecting" the shape of the density of states and localization length

R. Bosisio, C. Gorini, G. Fleury, & J.-L. Pichard, *New J. Phys*. 16:095005 (2014)

Inelastic regime: theory

Theory:

Percolation approach to solve the RRN:
Ambegaokar, Halperin, Langer → conductance (1971)
Zvyagin → thermopower (1973)
thermopower = energy averaged over the percolating path

Integrate between (μ-Δ; μ+Δ)

$$S = \frac{\langle E-\mu\rangle}{eT} = \frac{1}{eT}\,\frac{\int dE\,(E-\mu)\,\nu(E)\,p(E)}{\int dE\,\nu(E)\,p(E)}$$

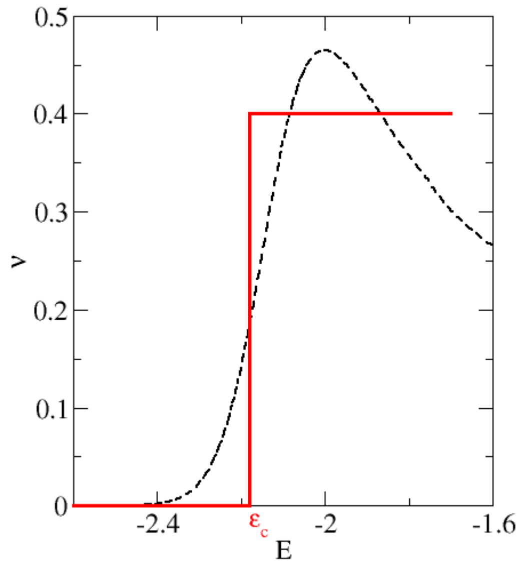
$\sim T^{-1}$: simple activation: energy to «jump» toward ϵ_C

$$S = \frac{k_B}{e}\left(\frac{\epsilon_c-\mu}{2k_BT} + \frac{\Delta(T)}{2k_BT}\right) \quad \text{if} \quad \epsilon_c < \mu$$ (inside)

$$S = \frac{k_B}{e}\left(\frac{\epsilon_c-\mu}{k_BT} + \frac{\Delta(T)}{2k_BT}\right) \quad \text{if} \quad \epsilon_c > \mu$$ (outside)

$\sim T^{-1/2}$: Variable Range Hopping (Mott)

$$\ln G(T) \sim -\frac{E_A}{k_BT} - \bar{\alpha}\sqrt{\frac{T_M}{T}}$$



Inelastic Regime: Typical thermopower

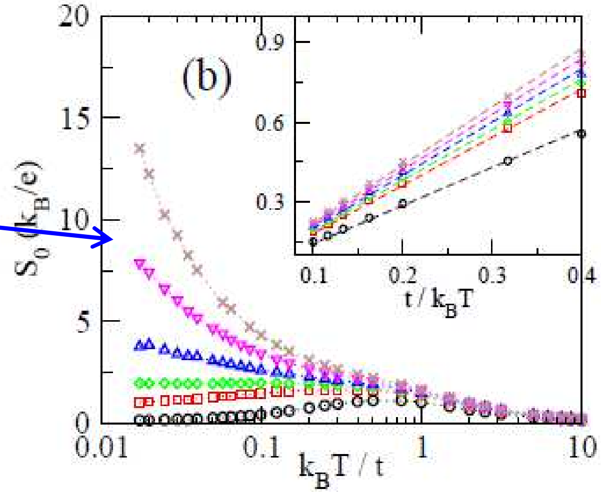
$$S = \frac{k_B}{e}\left(\frac{\epsilon_c - \mu}{2k_BT} + \frac{\Delta(T)}{2k_BT}\right) \quad \text{if} \quad \epsilon_c < \mu$$ (inside)

$$S = \frac{k_B}{e}\left(\frac{\epsilon_c - \mu}{k_BT} + \frac{\Delta(T)}{2k_BT}\right) \quad \text{if} \quad \epsilon_c > \mu$$ (outside)

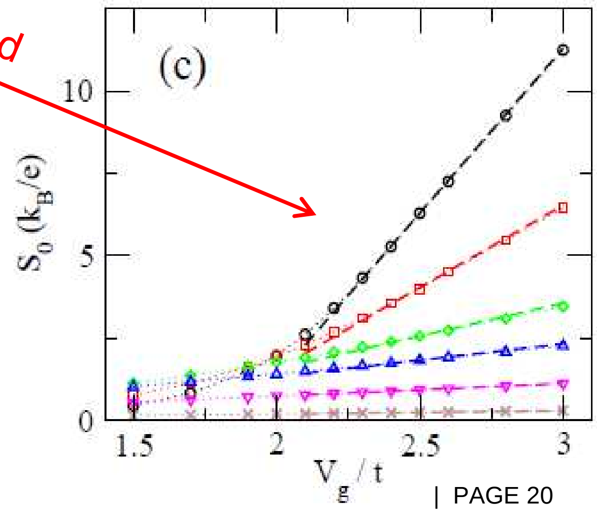
qualitative behavior of the slopes

linear behavior outside the band

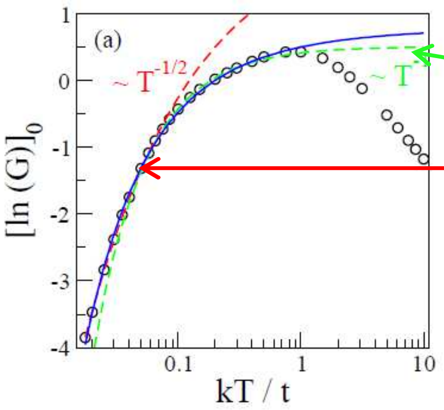
S_0 vs Temperature



S_0 vs Gate Voltage



Electrical Conductance



$$\ln G(T) \sim -\frac{E_A}{k_BT} - \alpha\sqrt{\frac{T_M}{T}}$$

R. Bosisio, C. Gorini, G. Fleury, & J.-L. Pichard, *New J. Phys*. 16:095005 (2014)

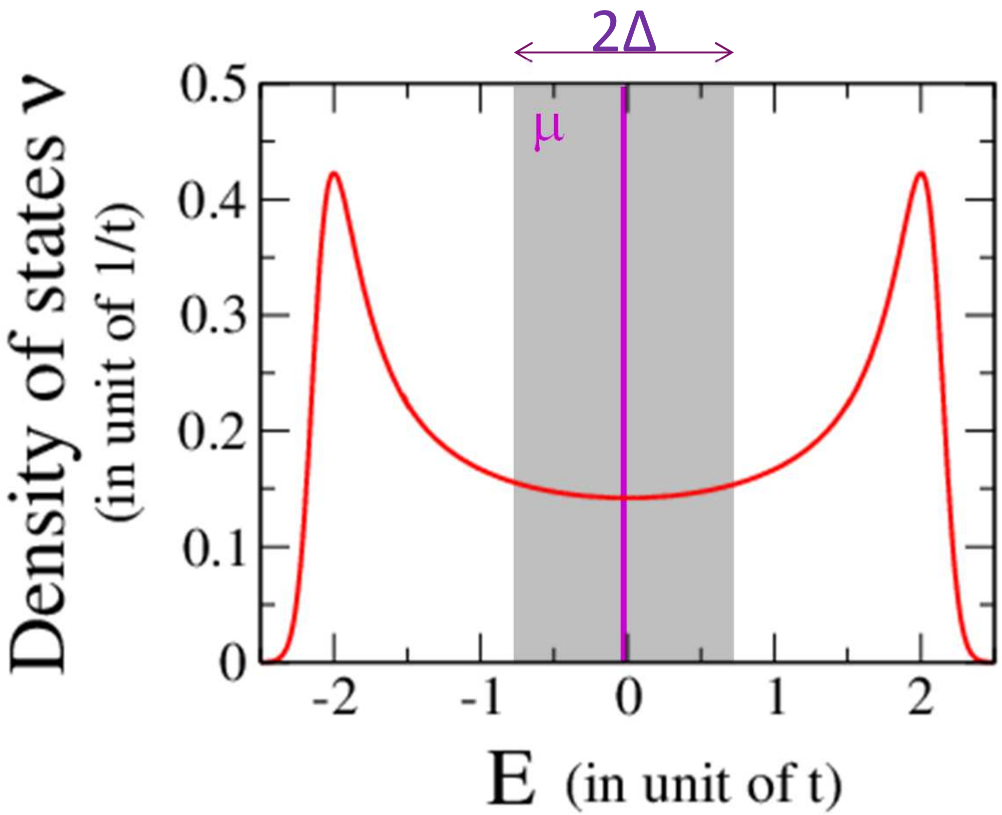
Inelastic Regime: Mott energy

$$S = \frac{\langle E-\mu\rangle}{eT} = \frac{1}{eT}\frac{\int dE\,(E-\mu)\,\nu(E)\,p(E)}{\int dE\,\nu(E)\,p(E)}$$

Integration inside [μ -Δ, μ +Δ]

Mott's Hopping Energy:
finite range of states contributing to transport

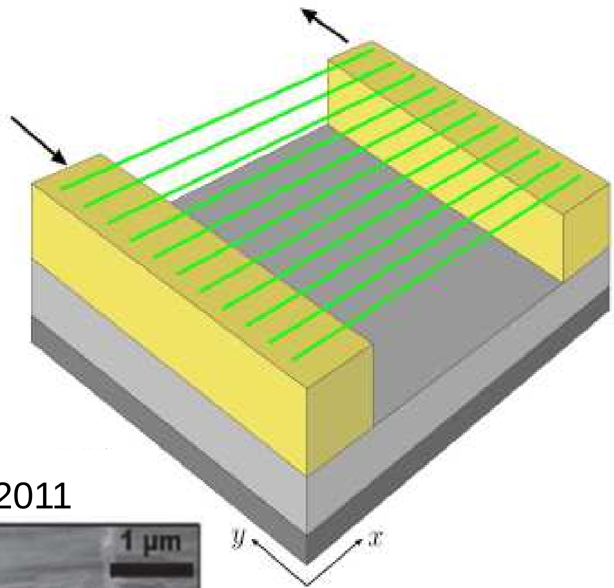
$$\Delta = k_B\sqrt{TT_M} \propto \sqrt{\frac{T}{\xi\nu}} \gg k_B T$$



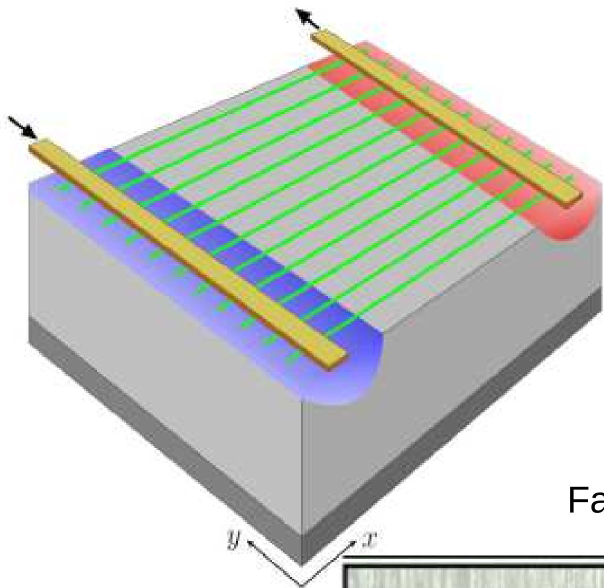
S depends on the asymmetry of the states around μ within [μ -Δ, μ +Δ]

3 - Arrays of parallel nanowires

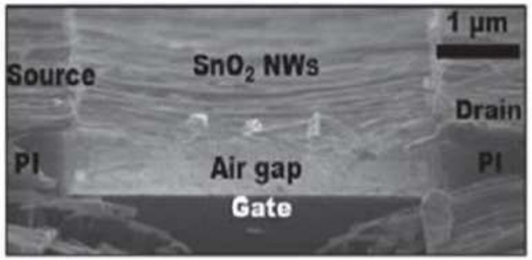
Suspended



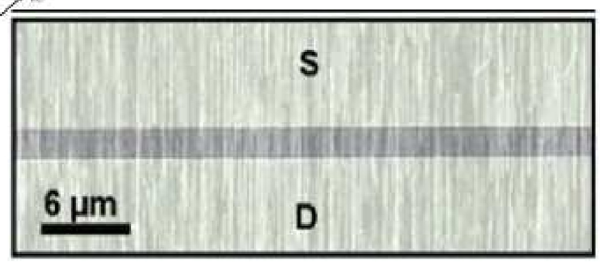
Deposited



Shin *et al.*, 2011



Fan *et al.*, 2008



- Neglect *inter*-wire hopping → independent nanowires
- Transport through each NW: VRH / NNH regime (same treatment as before)

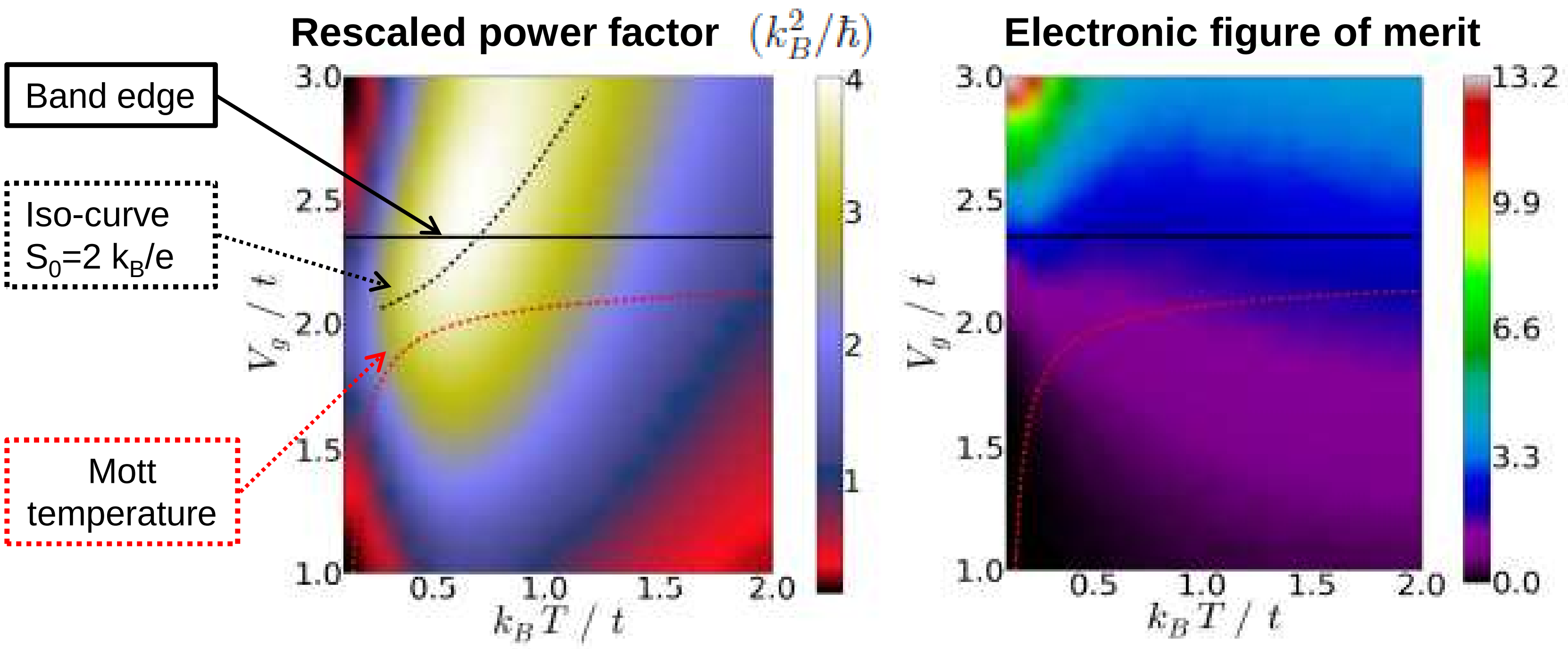
Parallel nanowires: power factor and figure of merit

$$G \approx M\, G_0$$
$$K^e \approx M\, K_0^e$$
$$S \approx S_0$$

Scalable Power Factor

(without affecting the electronic figure of merit)

$$Q = GS^2 \approx M\, G_0 S_0^2$$



Parameters: $M = 150,\ W = t,\ \gamma_e = \gamma_{ep} = t,\ L = 450$

P~μW for 10^5 NWs (1 cm) and δT~10 K

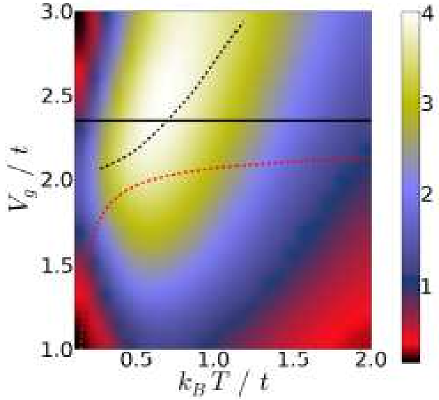
R. Bosisio, C. Gorini, G. Fleury, & J.-L. Pichard, *submitted to Phys. Rev. Appl.* (2014)

Parallel nanowires: power factor and figure of merit

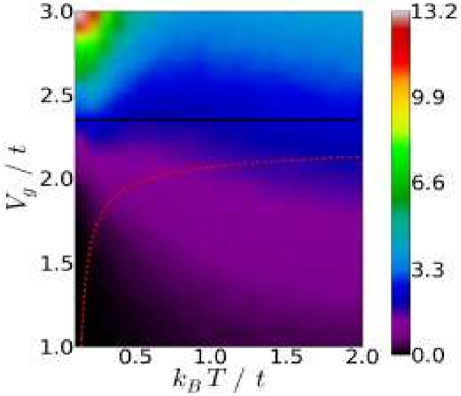
Estimation of the parasitic **phononic contribution** to ZT

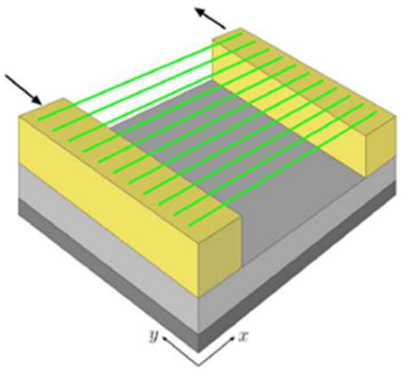
$$ZT = \frac{Z_e T}{1 + K^{ph}/K^e}$$

Rescaled power factor

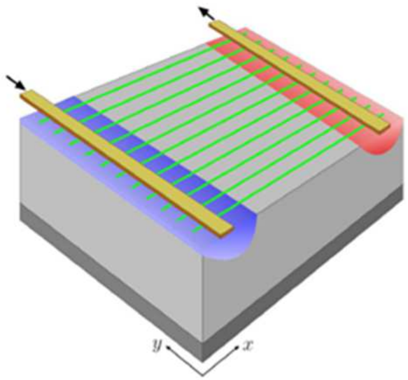


Electronic figure of merit





$$ZT \approx \frac{Z_e T}{3}$$



$$ZT \approx \frac{Z_e T}{21}$$

(For doped Si-NWs and SiO_2 substrate)

Parallel nanowires: Hot Spot cooling

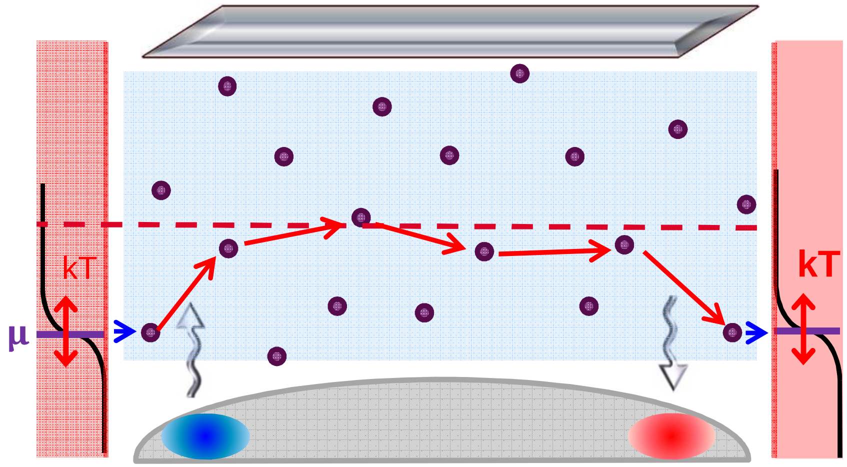
Hopping heat current through each localized state *i*

$$I_i^Q = \sum_j I_{ij}^Q = \sum_j (E_j - E_i) I_{ij}^N$$

E_i randomly distributed → Local fluctuations

$$\mathcal{I}_{x,y}^Q = \sum_{i \in \Lambda_{ph} \times \Lambda_{ph}(x,y)} I_i^Q$$

Λ_{ph}: inelastic phonon mean free path = *thermalization length* in the substrate



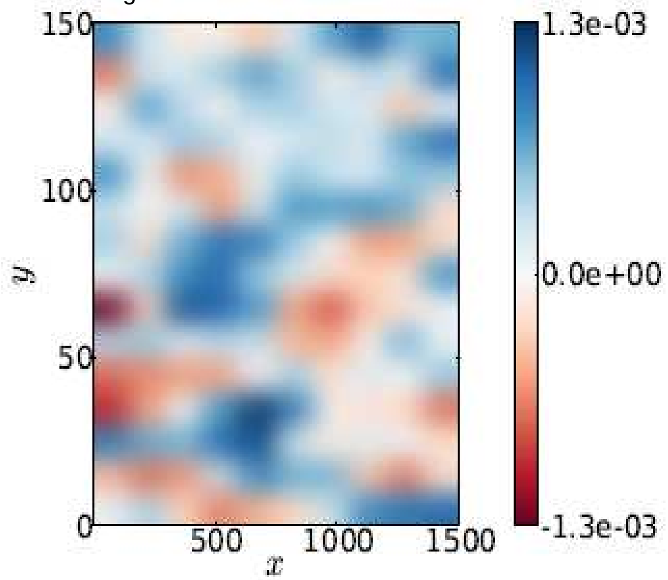
R. Bosisio, C. Gorini, G. Fleury, & J.-L. Pichard, *submitted to Phys. Rev. Appl.* (2014)

Parallel nanowires: Hot Spot cooling

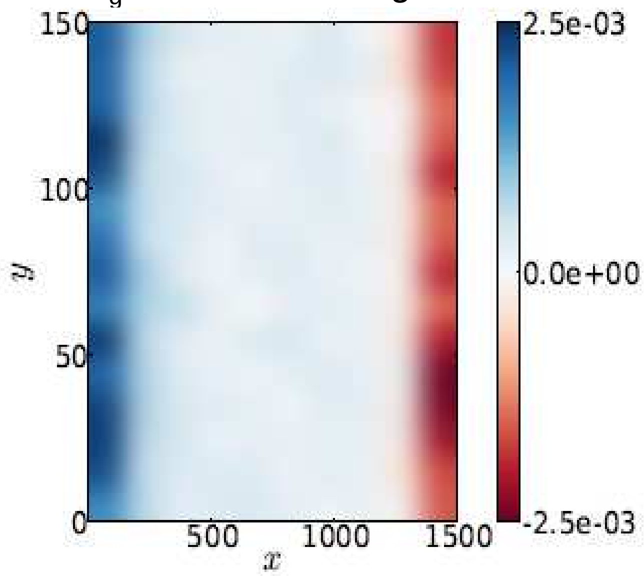
Top view of the substrate

Probing the lower (upper) band edge

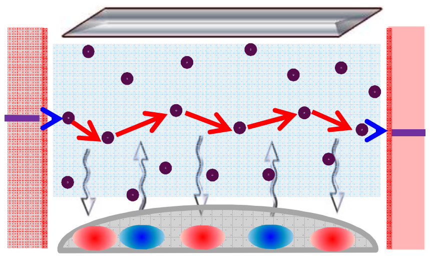
V_g=0: band *center*

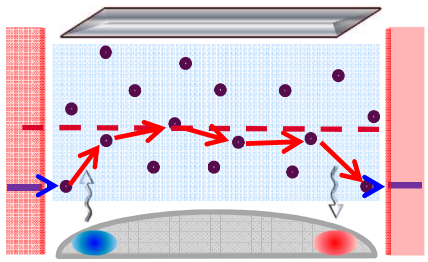


V_g=2.25 t: band *edge*



Formations of
a cold (hot) strip near
the source electrode
and of
a hot (cold) strip near
the drain electrode,





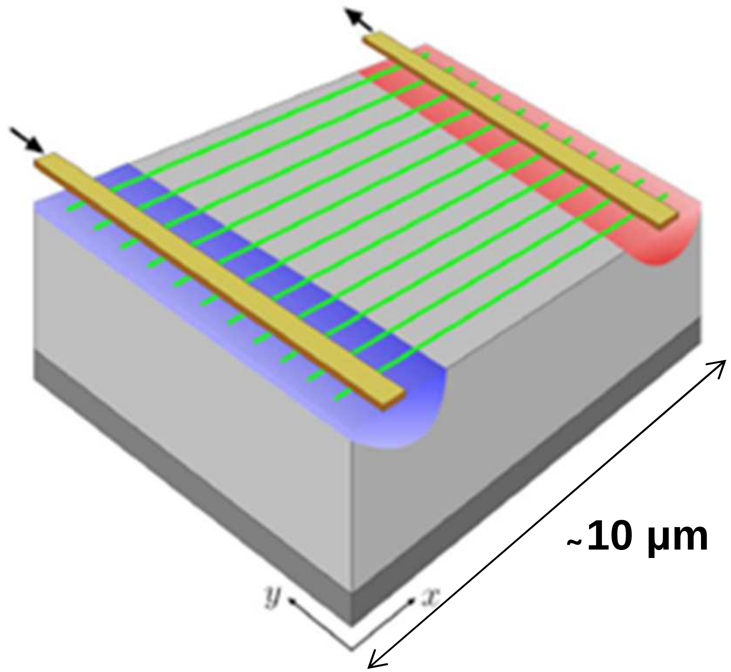
Parameters: $M = 150,\ W = t,\ \gamma_e = \gamma_{ep} = t,\ k_B T = 0.25t,\ \Lambda_{ph} = 150a$ (Heat currents in unit of $t^2/\hbar$)

R. Bosisio, C. Gorini, G. Fleury, & J.-L. Pichard, *submitted to Phys. Rev. Appl.* (2014)

Parallel nanowires: Hot Spot cooling

Opportunities for a gate control of heat in microstructures

E.g.: **hot spot cooling** in microelectronics: transferring heat some microns away



Estimate of the cooling power density:

~10 W/cm^2

at δμ=1mV ~ 7.7 x 10^{-2} t, T = 75K ~ 0.5t (linear regime)

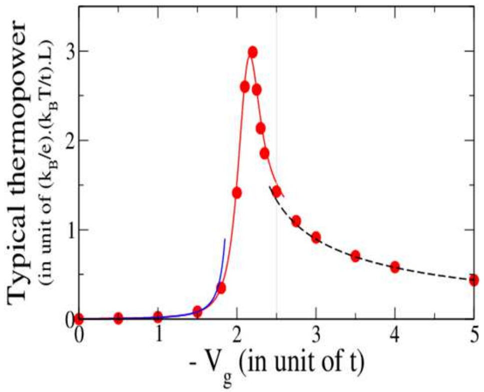
Electrodes of 1 cm long connected via 2.10^5 NWs

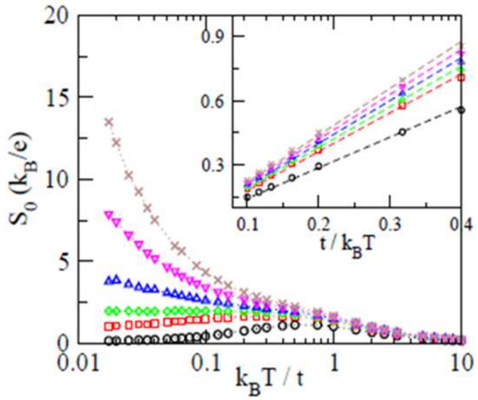
Strips of 1cm x 0.25 μm; T=77 K; $\delta V = 1$ V
0.15 W taken from the cold strip towards the hot strip located at 10 μm away

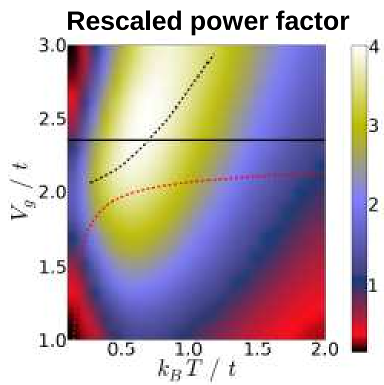
R. Bosisio, C. Gorini, G. Fleury, & J.-L. Pichard, *submitted to Phys. Rev. Appl.* (2014)

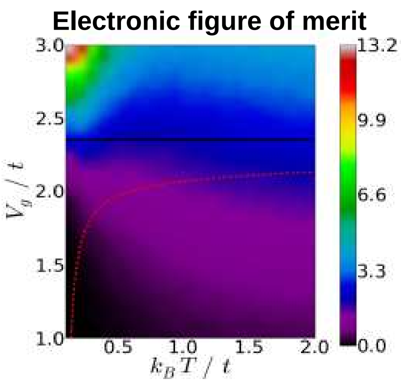
Summary

- Thermopower enhancement in disordered SC nws: low T and VRH
  - Opportunities offered by scalable modules

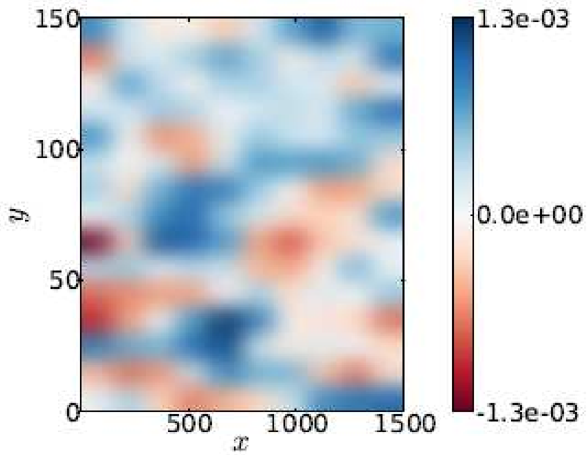


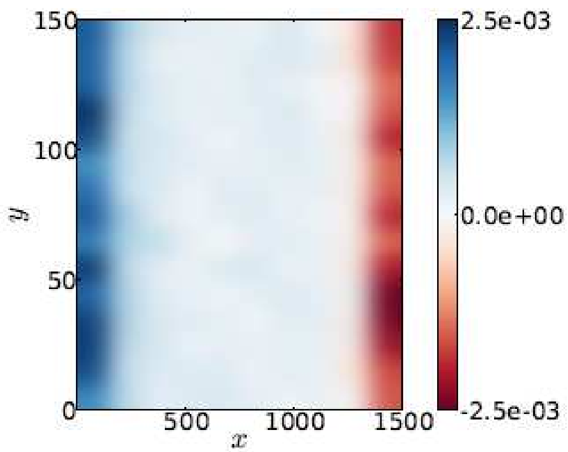






- Field control of phonon emission/absorption: Hot spot cooling





GMT group summer 2013

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Riccardo Bosisio
PhD Thesis

Thank You