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Thermoelectric Properties of Semiconductor Nanostructures

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EP3

Onsager Coefficients

- I electric current density
- J particle current density
- J_Q heat flux, heat current density
- μ chemical potential
- T temperature
- V voltage, electrostatic potential difference

$$\begin{pmatrix} I/-e \\ J_Q \end{pmatrix} = \begin{pmatrix} J \\ J_Q \end{pmatrix} = \begin{pmatrix} -\frac{L_{11}}{T} & -\frac{L_{12}}{T^2} \\ -\frac{L_{21}}{T} & -\frac{L_{22}}{T^2} \end{pmatrix} \begin{pmatrix} \nabla\mu - e\nabla V \\ \nabla T \end{pmatrix}$$

From: R.D. Barnard *Thermoelectricity in Metals and Alloys* (1972)

$$L_{11} = \frac{\sigma T}{e^2}$$

$$L_{12} = L_{21}$$

$$L_{12} = -\frac{ST^2\sigma}{e} = -\frac{\Pi T\sigma}{e}$$

$$L_{22} = T^2\left(\kappa + T\sigma S^2\right)$$

Thermoelectric Properties

"fluxes" $$\begin{pmatrix} I \\ Q \end{pmatrix} = \begin{pmatrix} G & L \\ M & K \end{pmatrix} \begin{pmatrix} \Delta\mu / e \\ \Delta T \end{pmatrix}$$ "forces"

Onsager-relation: $M = -LT$

$$\begin{pmatrix} -\Delta V \\ Q \end{pmatrix} = \begin{pmatrix} R & S \\ \Pi & -\kappa \end{pmatrix} \begin{pmatrix} I \\ \Delta T \end{pmatrix}$$

Diffusion Thermopower

$$S \equiv \left(\frac{\Delta\mu / e}{\Delta T} \right)_{I=0} = -\frac{L}{G}$$

$$\Pi \equiv \left(\frac{Q}{I} \right)_{\Delta T=0} = \frac{M}{G} = ST$$

$$\kappa \equiv -\left(\frac{Q}{\Delta T} \right)_{I=0} = -K\left(1 + \frac{S^2 GT}{K} \right)$$

Thermoelectric Properties

Landauer-Büttiker-Formalism:

$$G = -\frac{2e^2}{h}\int_0^\infty dE\,\frac{\partial f}{\partial E}t(E)$$

$$L = -\frac{2e^2}{h}\frac{k_B}{e}\int_0^\infty dE\,\frac{\partial f}{\partial E}t(E)\frac{(E-E_F)}{k_B T}$$

$$\frac{(E-E_F)}{k_B T}\left(\frac{\partial f}{\partial E}\right)$$

odd function in *E*
➔ *L* large for *t(E)* asymmetric around E_F

$$\frac{K}{T} = \frac{2e^2}{h}\left(\frac{k_B}{e}\right)^2\int_0^\infty dE\,\frac{\partial f}{\partial E}t(E)\left[\frac{(E-E_F)}{k_B T}\right]^2$$

$$S \equiv \left(\frac{\Delta\mu/e}{\Delta T}\right)_{I=0} = -\frac{L}{G}$$

$$\Rightarrow S = -\frac{\langle E\rangle}{eT}$$

Thermopower (S)

- Kelvin-Onsager relation (1931)

$$S = -\frac{L}{G}\bigg|_{I=0} = \frac{\Pi}{T} = -\frac{\langle E \rangle}{eT}$$

$(\Delta Q = T\Delta \mathbf{S})$ thermal energy to transfer one electron from a hot to a cold reservoir

- Heike's formula

$$S = -\frac{1}{e}\Delta \mathbf{S} = -\frac{1}{e}k_B\left(\ln g_f - \ln g_i\right)$$

(spin) entropy contribution

- Mott relation

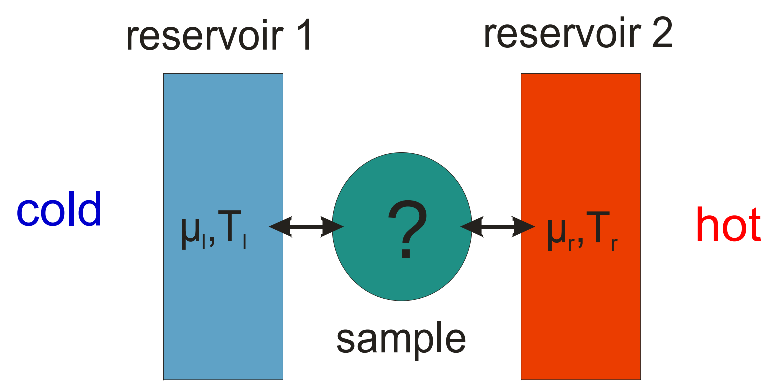
$$S = -\frac{\pi^2}{3}\frac{k_B}{q}\frac{k_B T}{G}\frac{dG}{dE}\bigg|_{E_F}$$

linear response

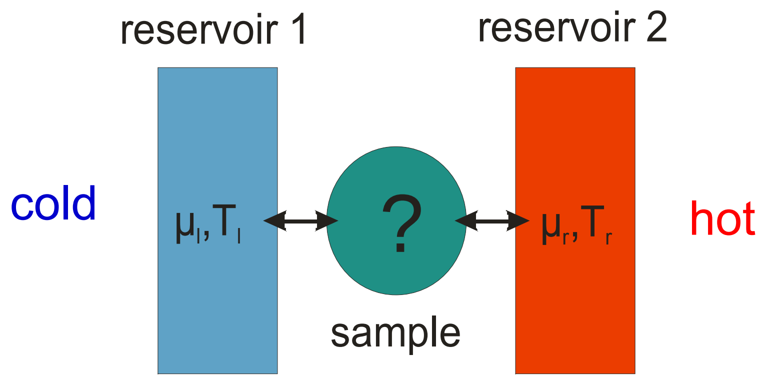
Thermopower (S)

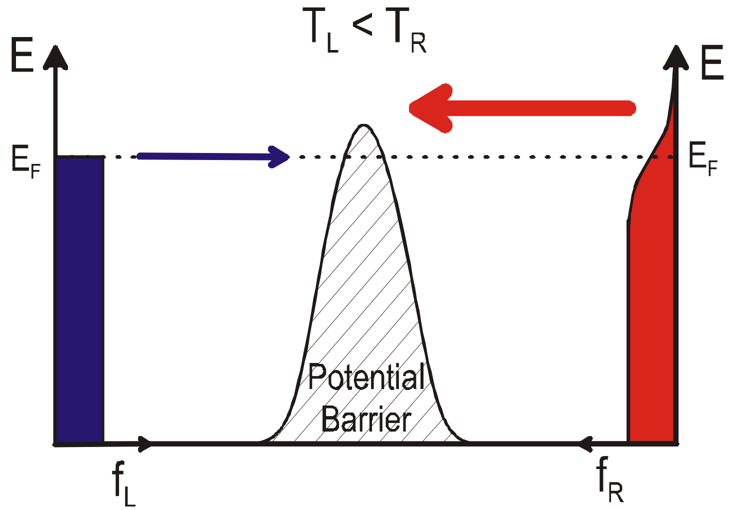
Measurement of the Thermopower

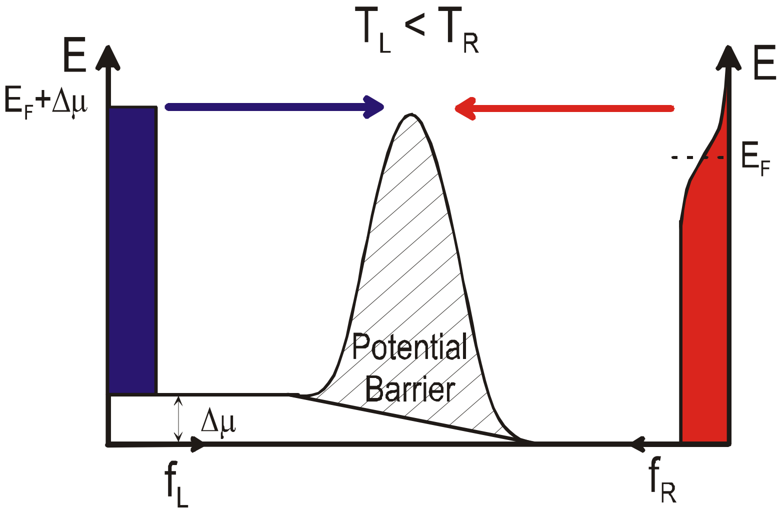
$$S \equiv - \lim_{\Delta T \to 0} \left. \frac{\Delta V_{th}}{\Delta T} \right|_{I=0}$$



Thermopower Measurement

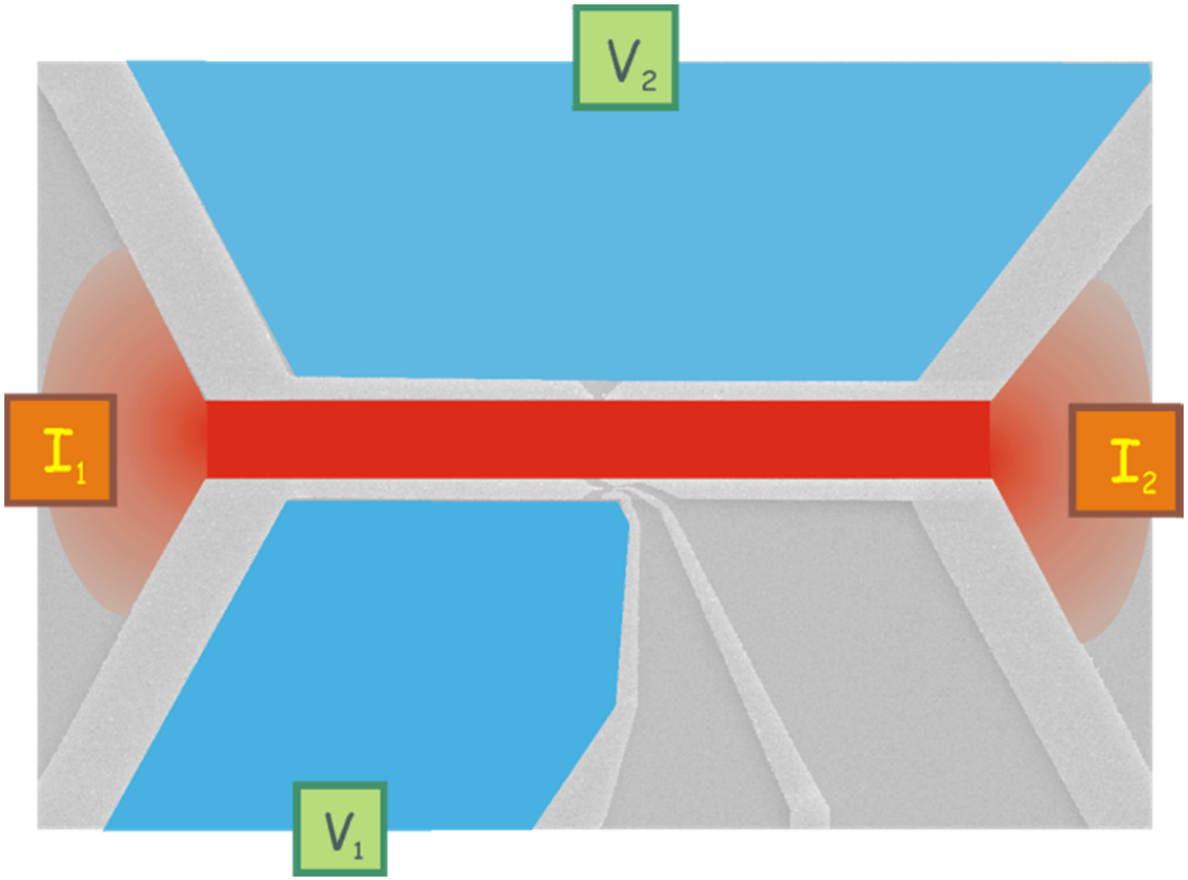






$$S := -\lim_{\Delta T \to 0} \frac{V_{th}}{\Delta T}\bigg|_{I=0}$$

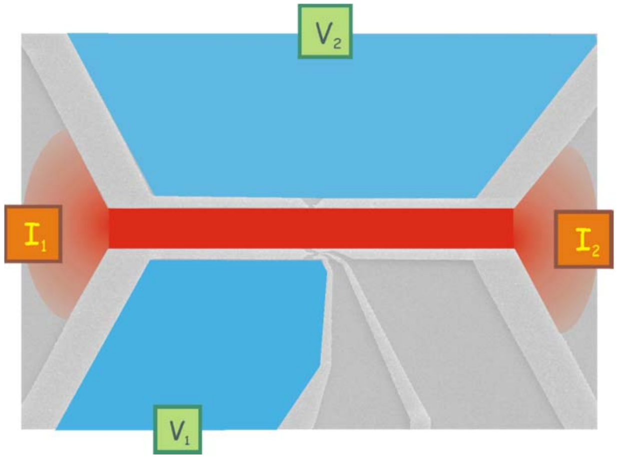
Current Heating Technique

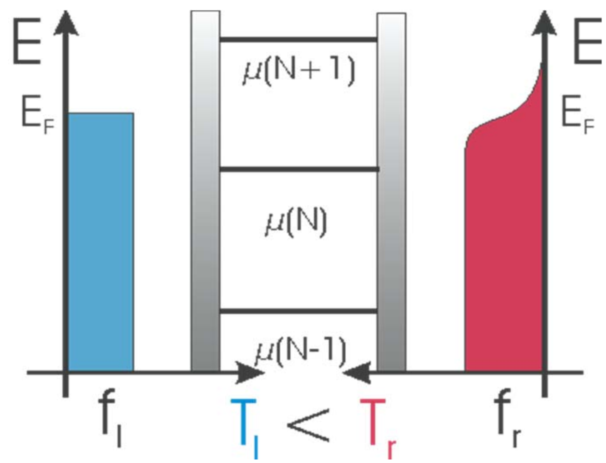


$$V_{th} = V_1 - V_2 = \left(S_{dot} - S_{qpc}\right)\left(T_e - T_L\right)$$

Current Heating Technique

$$V_{th} = V_1 - V_2 = (S_{dot} - S_{qpc})(T_e - T_L)$$





- energy dissipation at the channel entrance
- only hot electron gas within channel
  (1 ps ≈ τ_{ee} << τ_{eph} ≈ 0.2 ns)
- energy relaxation in the reservoir
- diffusion thermopower

  ΔT = 10 mK, Δx = 500 nm ➔ 20 K/mm

- QD and QPC create thermovoltages which can be measured as voltage difference between V_1 and V_2

  $V_1 - V_2 = (S_{QD} - S_{QPC})\,\Delta T = S_{QD}\,\Delta T$

  S_{QPC} can be adjusted to zero

- ac-excitation and detection:
  P_{heat} ~ $[I \sin(\omega t)]^2$
  ~ $\sin(2\omega t)$ ($\omega/2\pi = 13$ Hz)

First Experiments: Thermopower of a QPC

VOLUME 65, NUMBER 8 PHYSICAL REVIEW LETTERS 20 AUGUST 1990

Quantum Oscillations in the Transverse Voltage of a Channel in the Nonlinear Transport Regime

L. W. Molenkamp, H. van Houten, C. W. J. Beenakker, and R. Eppenga
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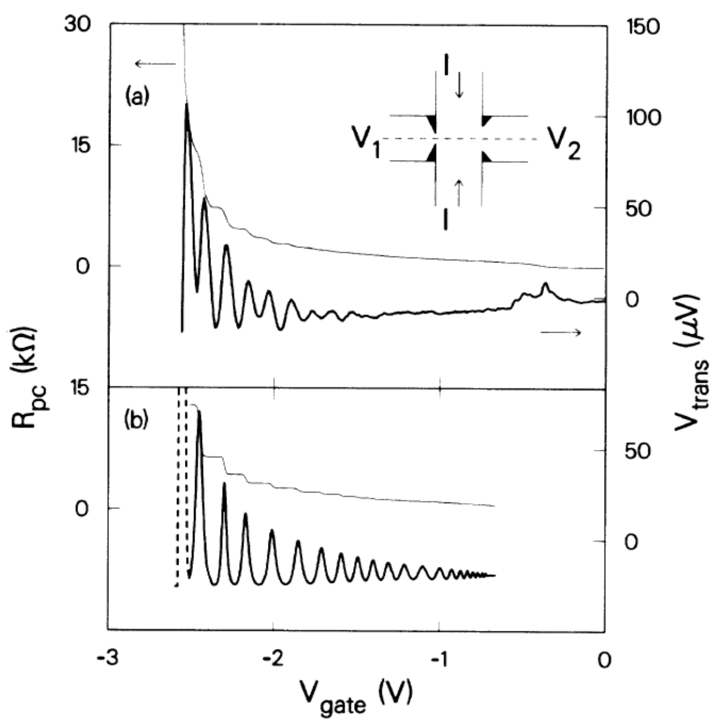
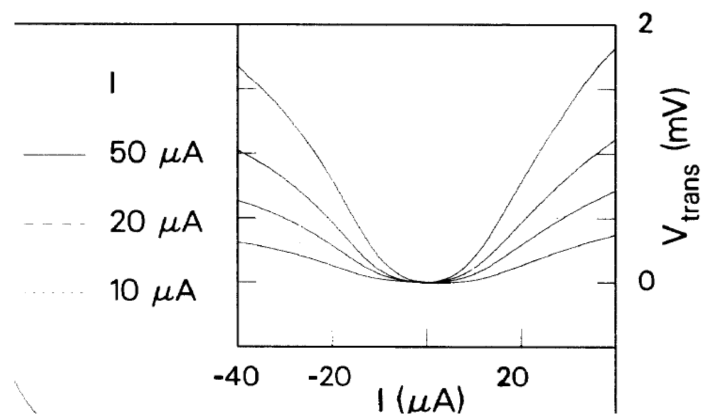
(Received 5 March 1990)

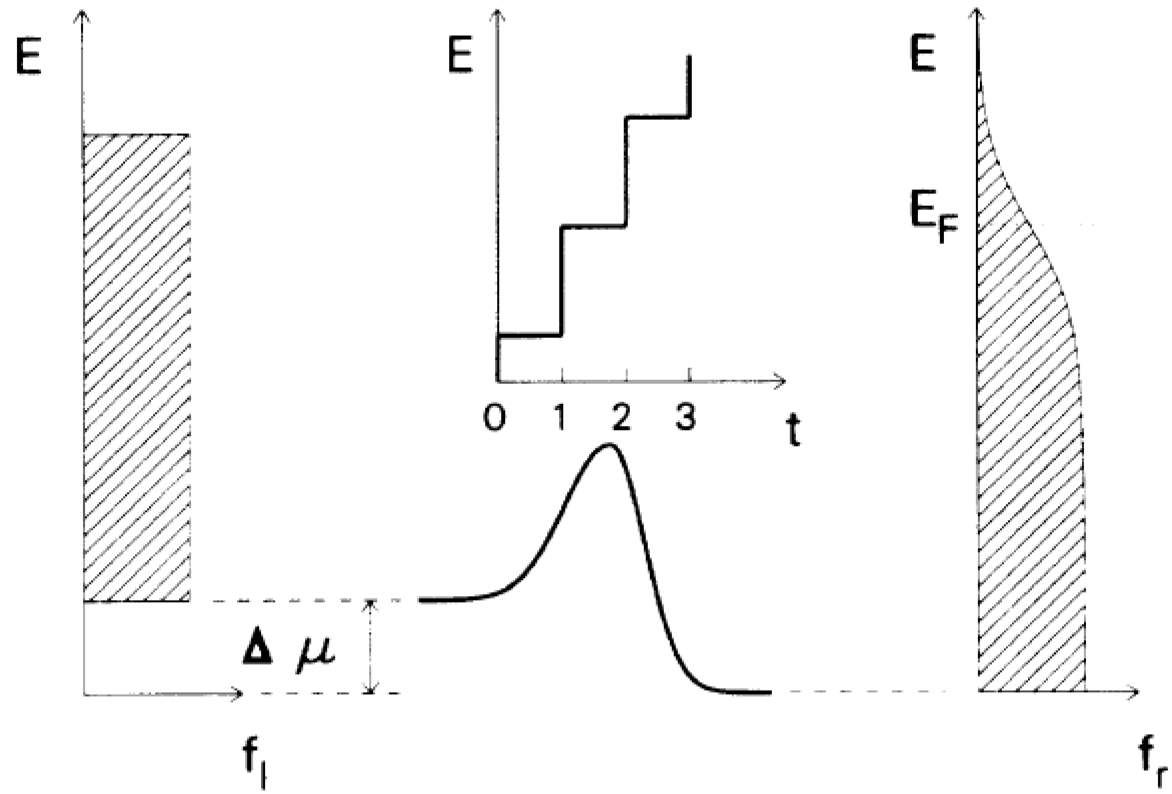




In semiconductors, at low T, $\tau_{e-p} \sim 100$ ps.

$\rightarrow$ nearly thermalized hot electron distribution in the heating channel

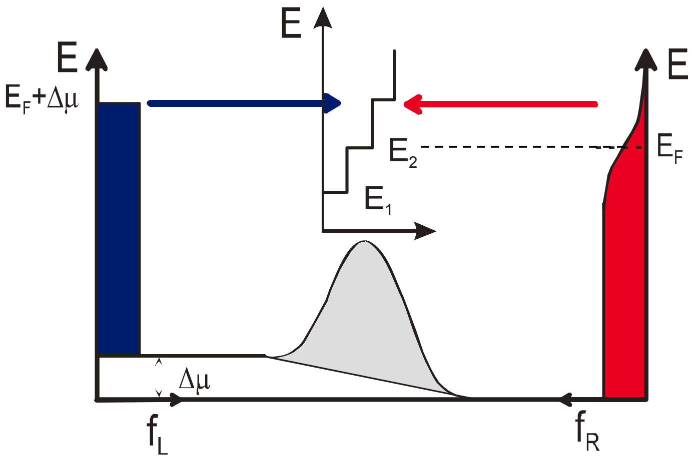
Step-by-step Barrier



Each channel in the point contact acts as a potential barrier, hence the thermopower shows a series of peaks

L.W. Molenkamp et al., Phys. Rev. Lett. 65, 1052 (1990).

Thermopower of a QPC



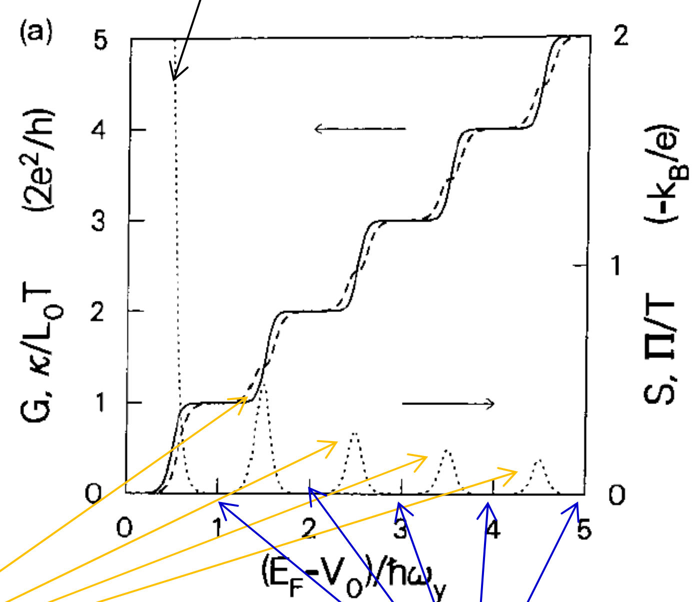
$$\int_0^\infty f dE = k_B T \ln\left[1+\exp\left(E_F / k_B T\right)\right]$$

$$\Rightarrow L = \frac{2e^2}{h}\frac{k_B}{e}\sum_{n=1}^{\infty}\left[\ln\left(1+e^{-\varepsilon_n}\right)+\ln\left(1+e^{\varepsilon_n}\right)^{-1}\right]$$

quantized thermopower

$$S = -\frac{k_B}{e}\left(1+\varepsilon_1\right) \quad \text{if} \quad N<1$$

H. van Houten et al., Semicond. Sci. Technol. **7**, B215 (1992)

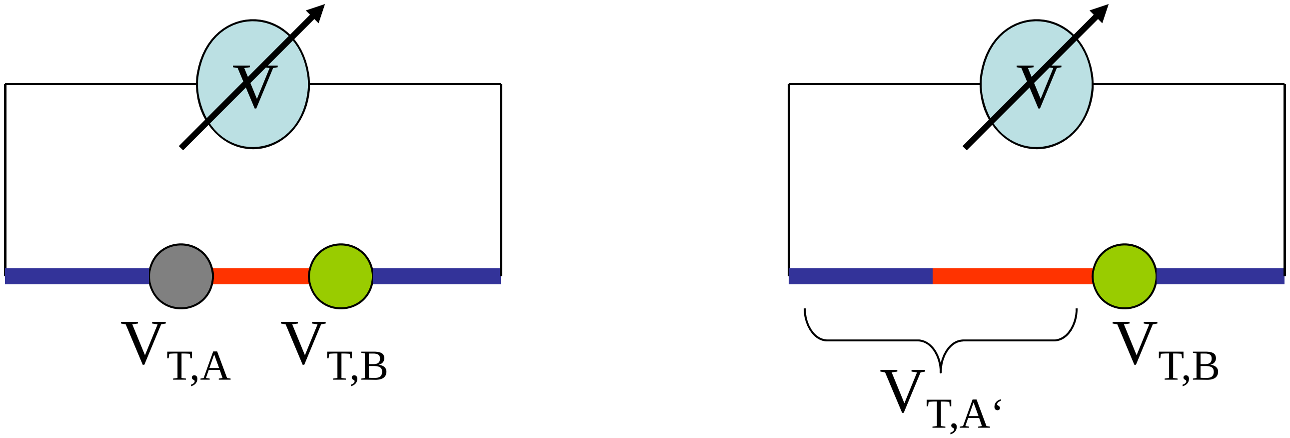


$$S = -\frac{k_B}{e}\frac{\ln 2}{N-\frac{1}{2}} \quad \text{if} \quad E_F = E_N; \quad N>1$$

$$S = 0 \quad \text{if} \quad E_F \neq E_N; \quad N>1$$

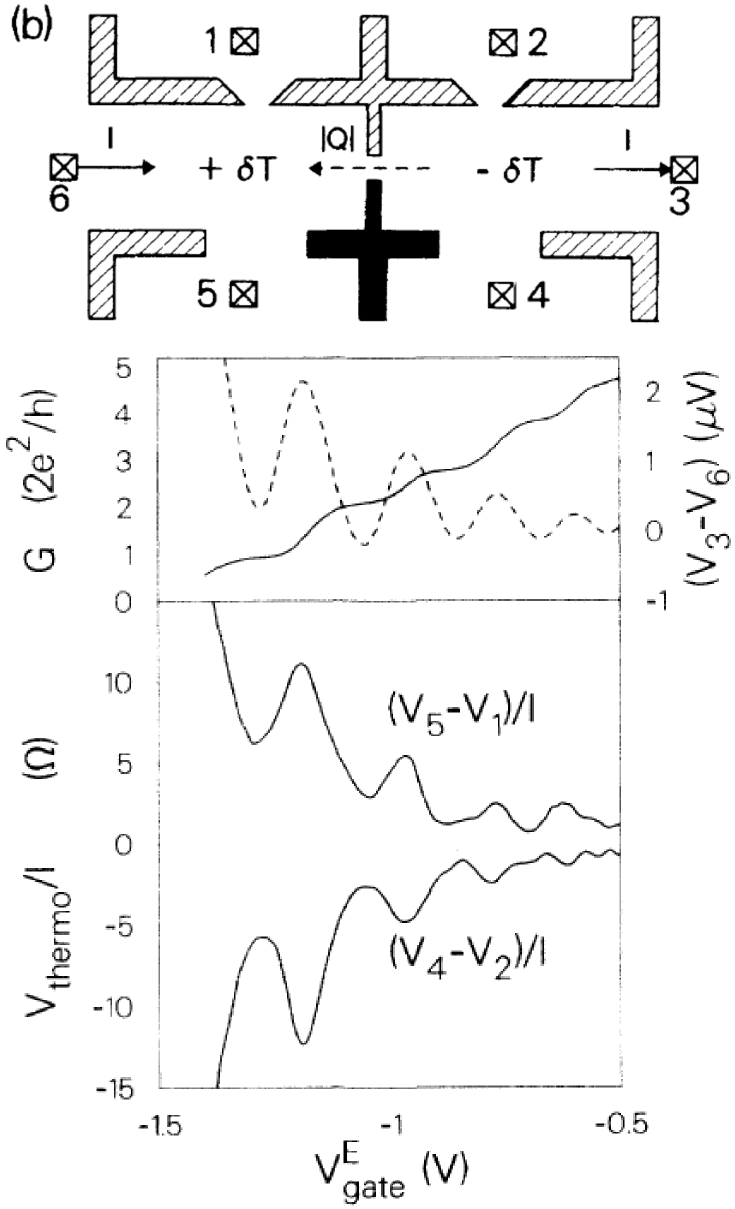
Reference QPC

- Voltage Probes have to be at same temperature and of the same material
- QPC can be used as a reference since TP of QPC is known (can be adjusted to zero)
- G of QPC is quantized – and therefore, so is S. This can be used as one method of temperature calibration



L.W. Molenkamp et al., Phys. Rev. Lett. 65, 1052 (1990).
L.W. Molenkamp et al., Phys. Rev. Lett. 68, 3765 (1992).
A.A.M. Staring et al., Europhys. Lett. 22, 57 (1993).
S. Möller et al., Phys. Rev. Lett. 81, 5197 (1998).
S.F. Godijn et al., Phys. Rev. Lett. 82, 2927 (1999).
R. Scheibner et al., Phys. Rev. Lett. 95, 176602 (2005).
R. Scheibner et al., Phys. Rev. B75, 041301(R) (2007).

Peltier Coefficient



Kelvin-Onsager relation $\Pi = ST$

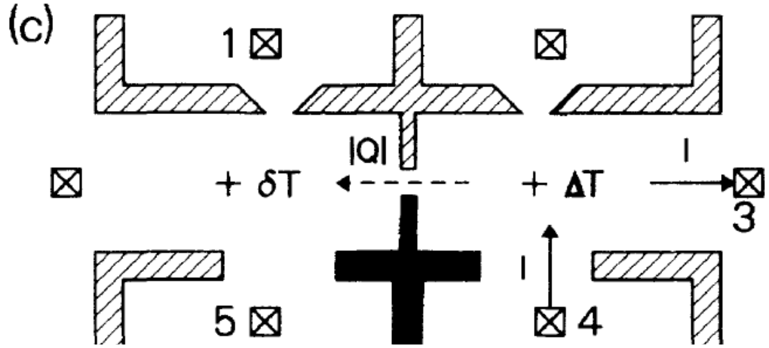
Theoretical estimate for Peltier coefficient

$$\Pi = ST = -(k_B T \ln 2)/(N + \tfrac{1}{2})e \approx -70\ \mu\text{V}$$

is within factor of 2 from observed signal.

L.W. Molenkamp et al., Phys. Rev. Lett. 68, 3765 (1992).

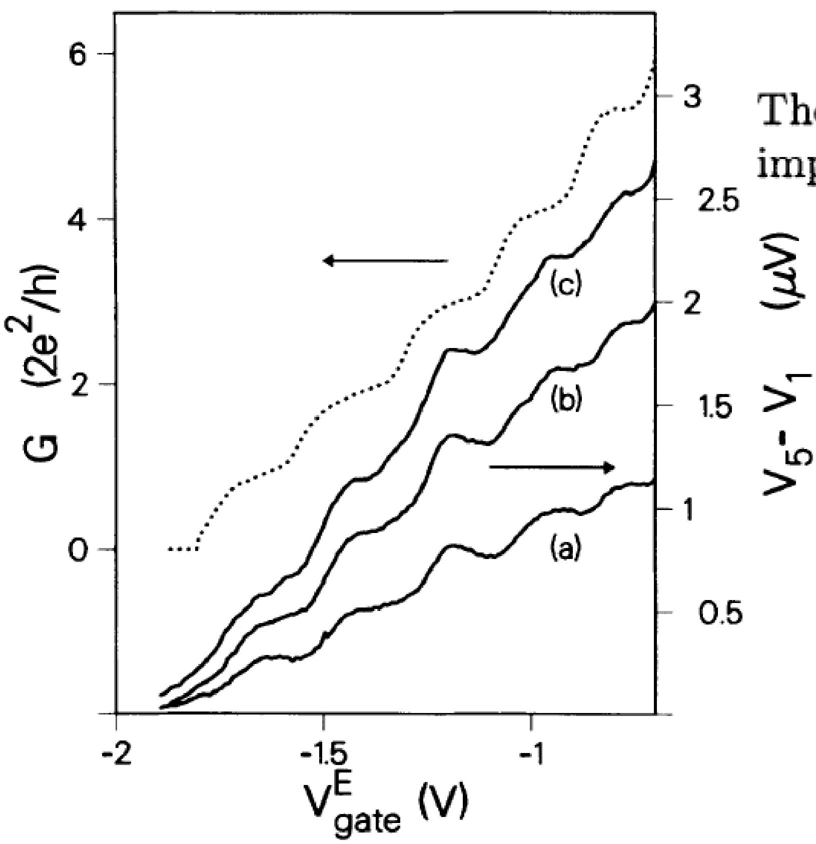
Thermal Conductance



Wiedemann- Franz relation,

$$\kappa \approx L_0 T G \ ,$$

$L_0 \equiv k_B^2 \pi^2 / 3e^2$ is the Lorenz number.



The Wiedemann-Franz relation (5), using $G = N(2e^2/h)$, implies $\kappa = 1.7 \times 10^{-11}$ W/K (for the $N = 5$ plateau).

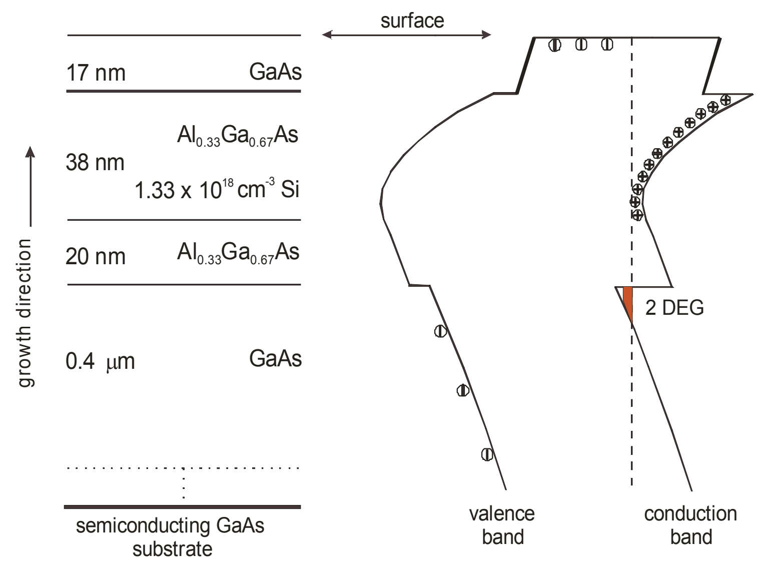
again within factor of 2 from the observed signal.

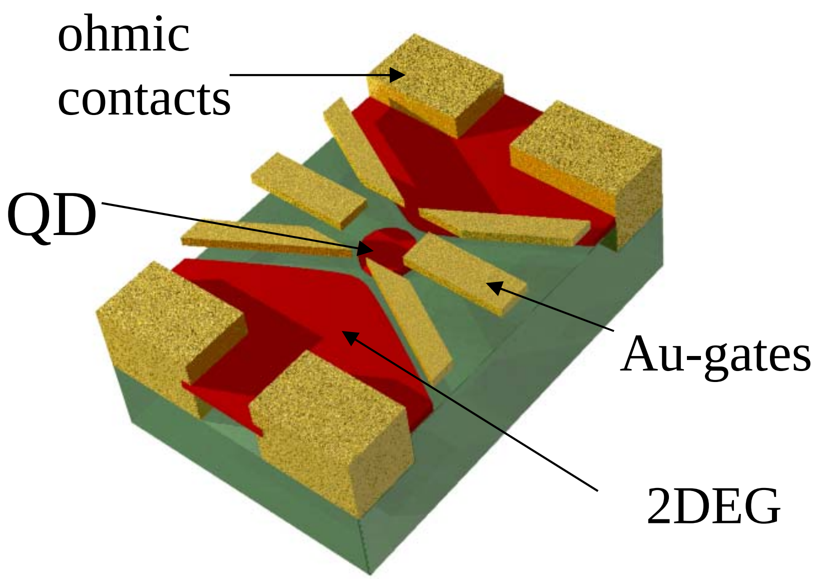
Wiedemann-Franz yields thermal conductance quantum.

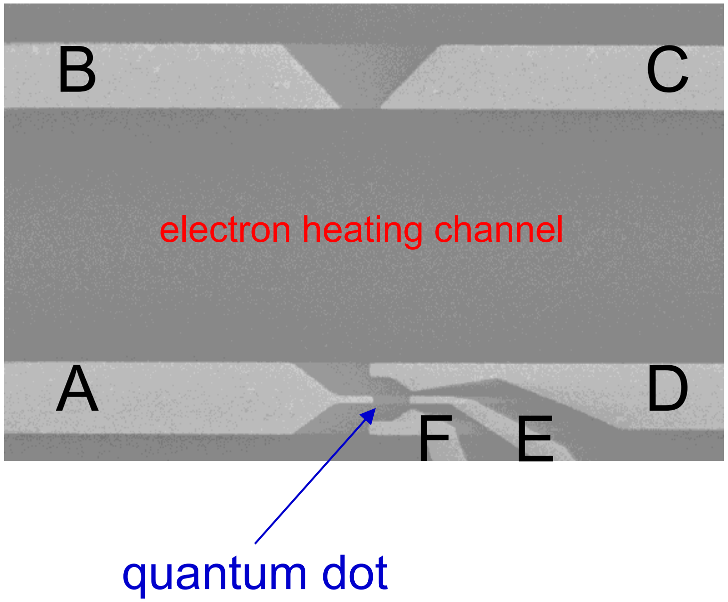
L.W. Molenkamp et al., Phys. Rev. Lett. 68, 3765 (1992).

Next: quantum dots

- GaAs/AlGaAs - 2DEG
- n = 2.3 10^{11} cm^{-2}, μ = 10^6 cm^2/Vs
- Ti/Au-surface electrodes
- (opt. and e-beam lithography)
- Au/AuGe - ohmic contacts

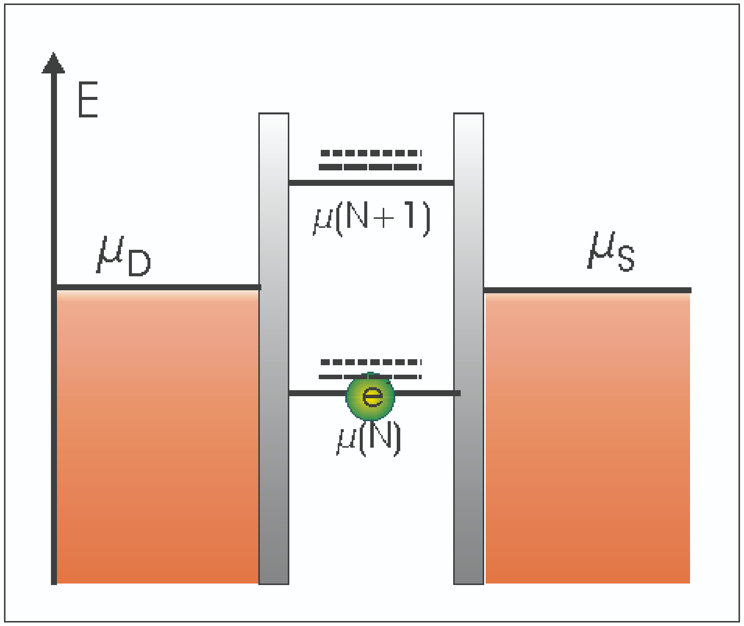






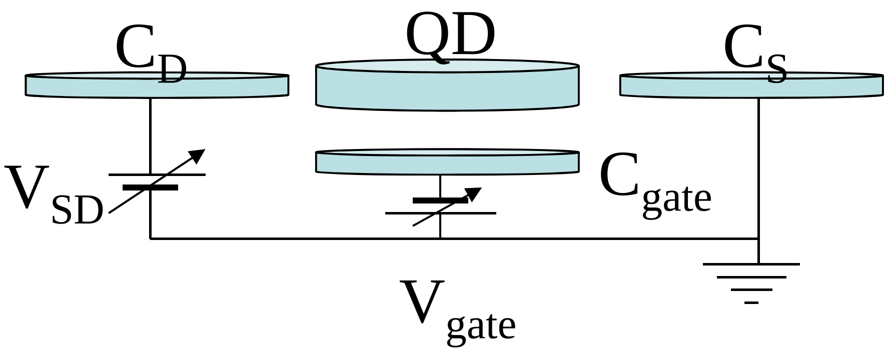
Quantum Dot (QD)

- Constant Interaction model:
  - QD = small capacitor
  - energies depend linearly on V_{gate}
  - coefficients do not depend on N (number of electrons)

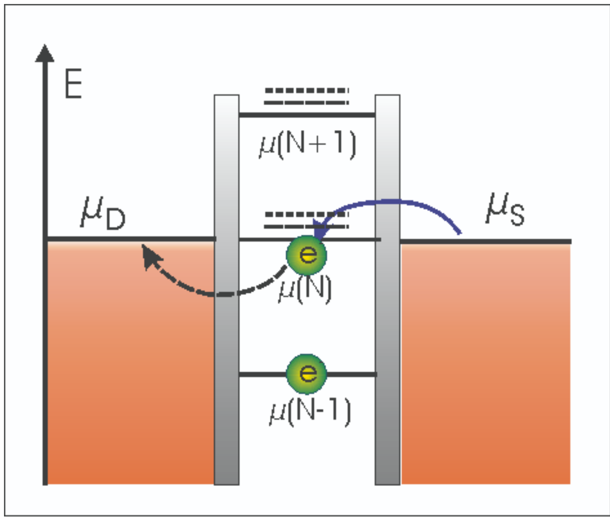


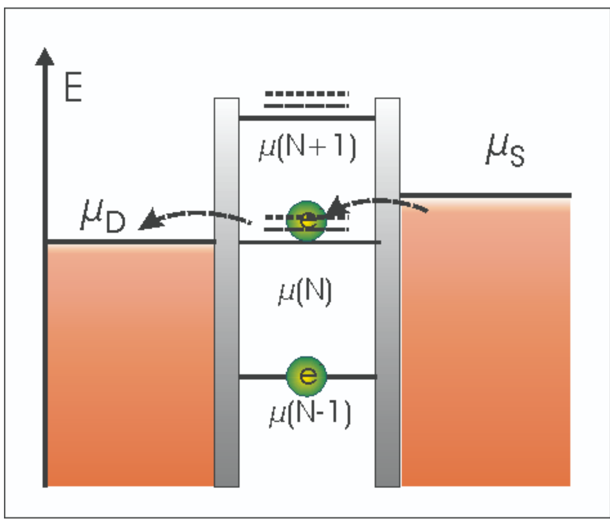
- Energy needed to add one electron:
  - qm. Energy E_{qm} ~ 100 µeV
  - Coulomb Interaction $E_C = ½\, e^2/C$ ~ 2 meV
  - $E_C = E_{qm} + E_C$

- Parameters accessible in conventional transport experiments

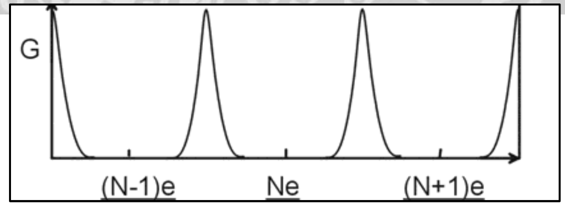


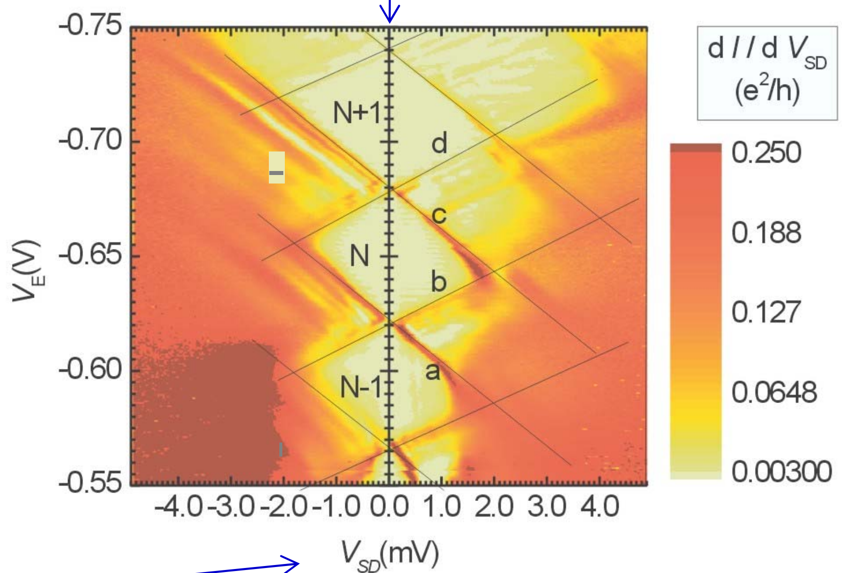
Transport Properties

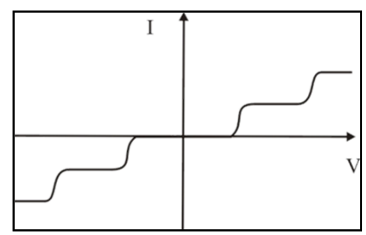




linear transport





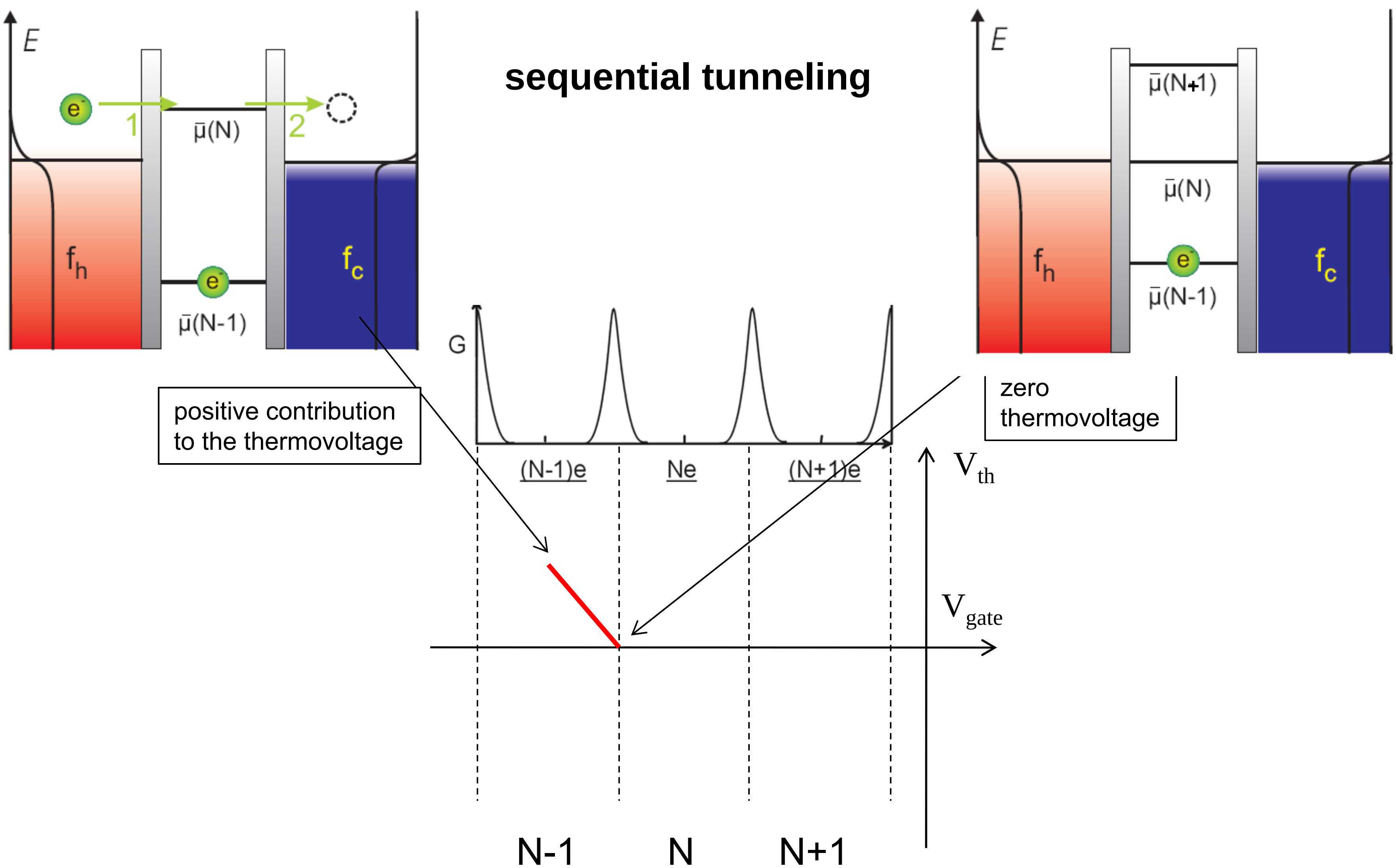


non-linear transport:

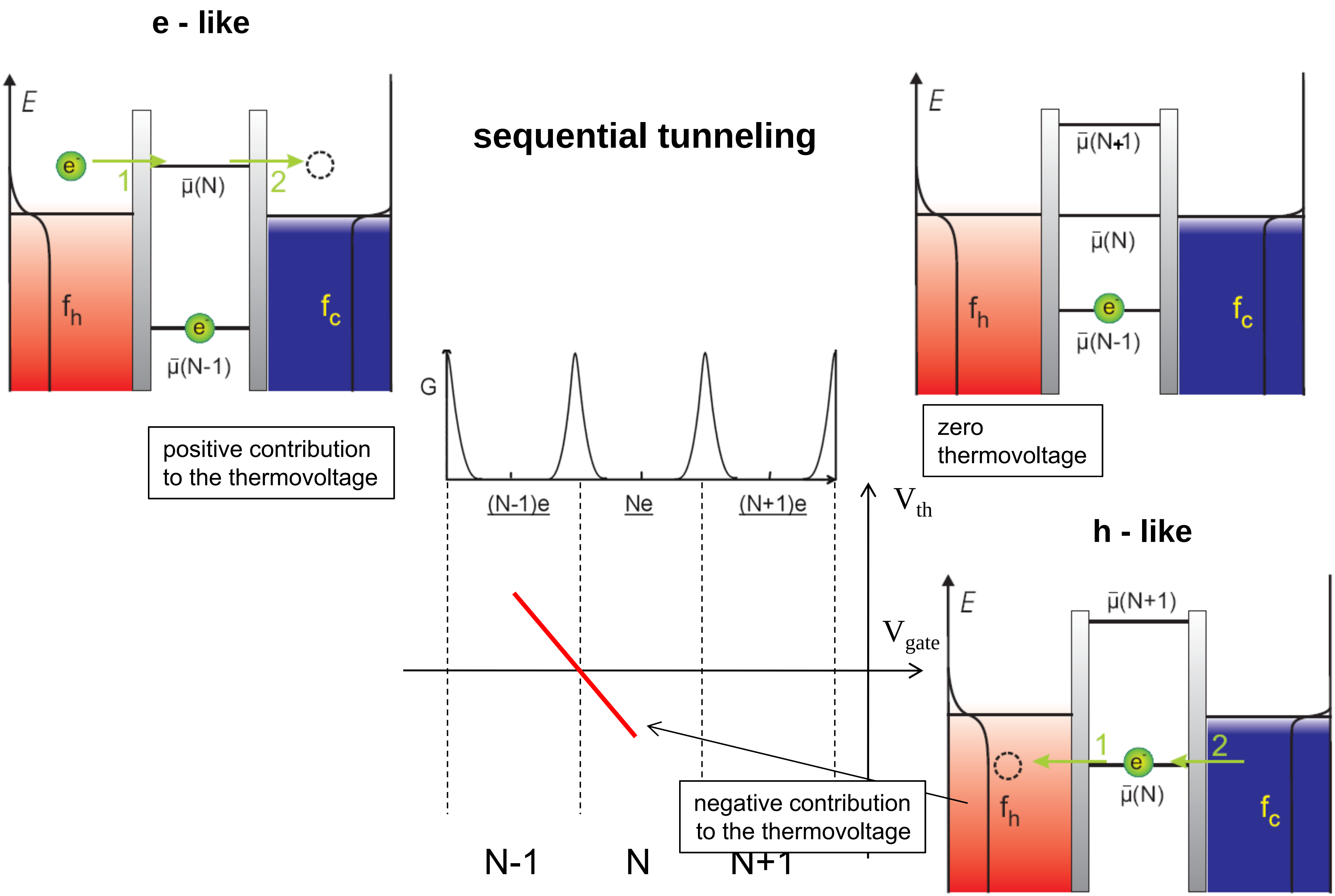
- capacitive coupling of leads and QD
- strong influence on hybridization of leads and QD

Thermopower of a QD

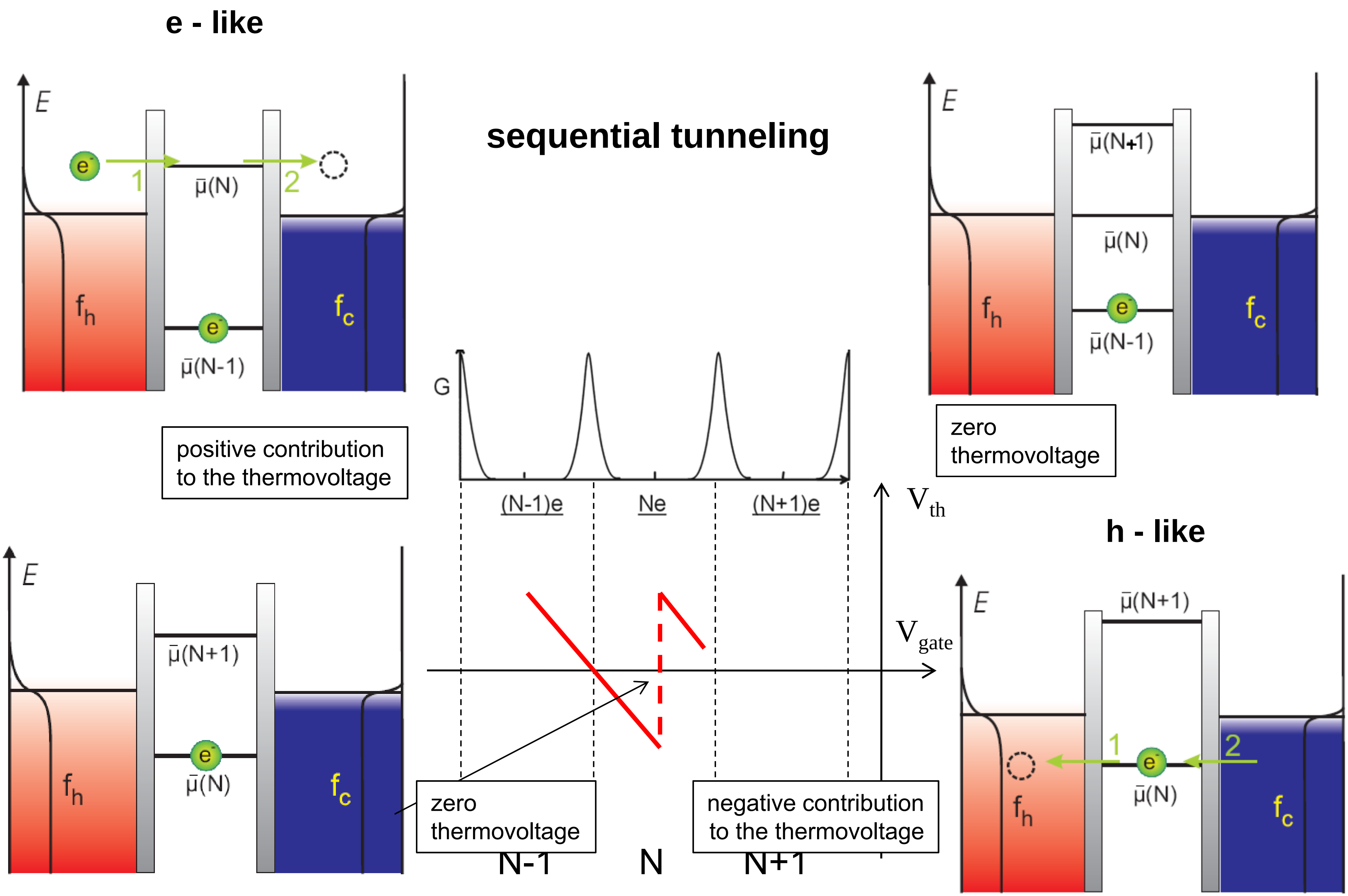
e - like



Thermopower of a QD



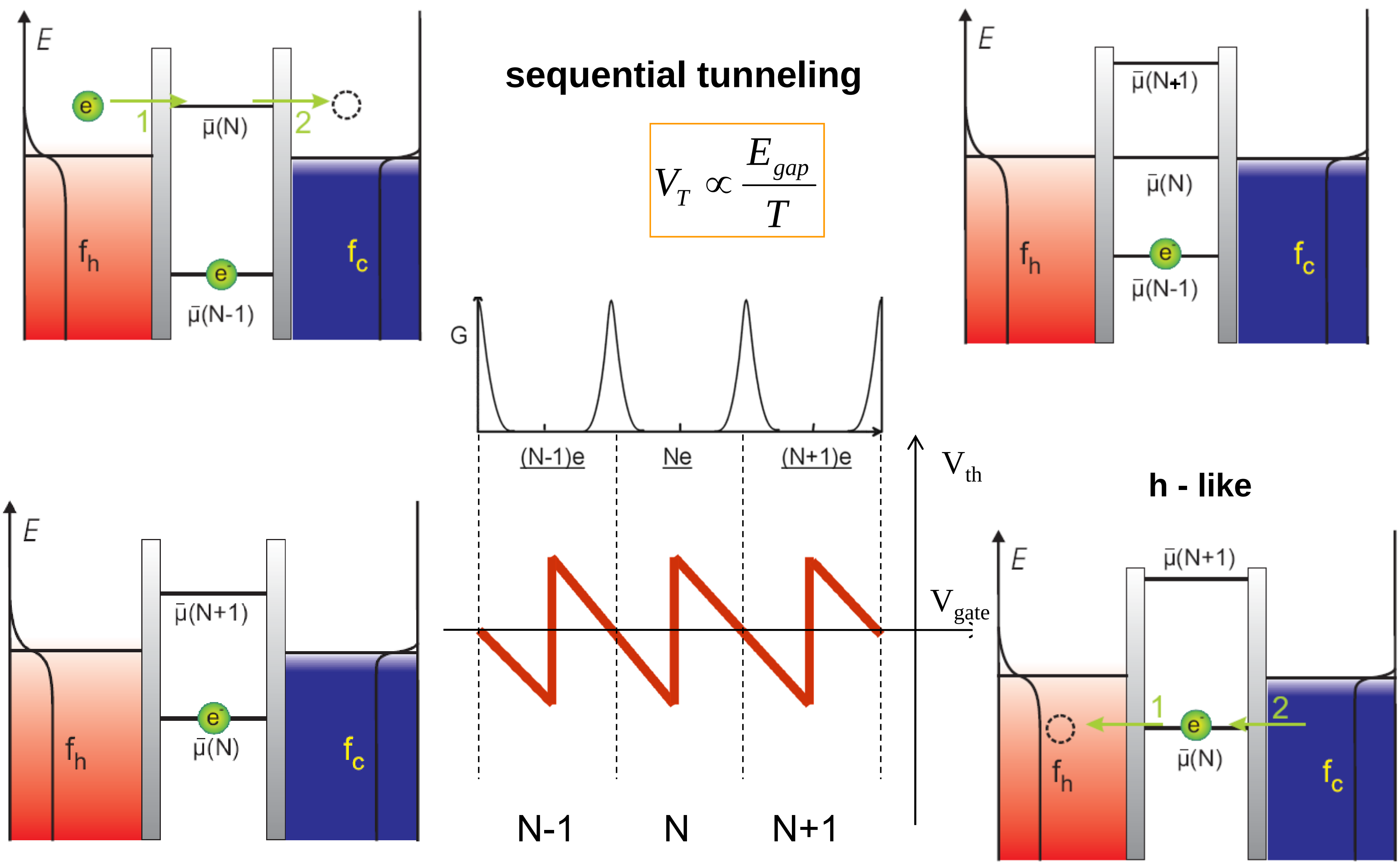
Thermopower of a QD



Thermopower of a QD

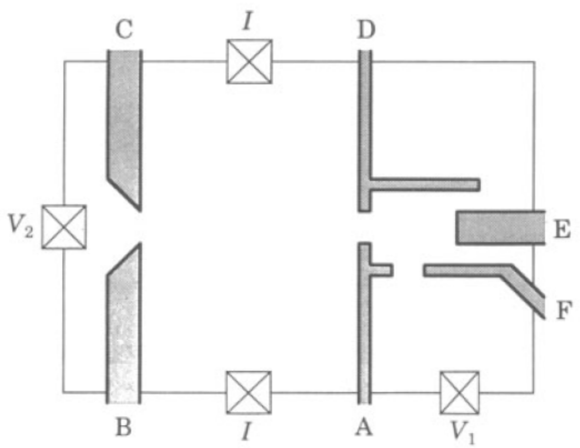
sequential tunneling

$$V_T \propto \frac{E_{gap}}{T}$$

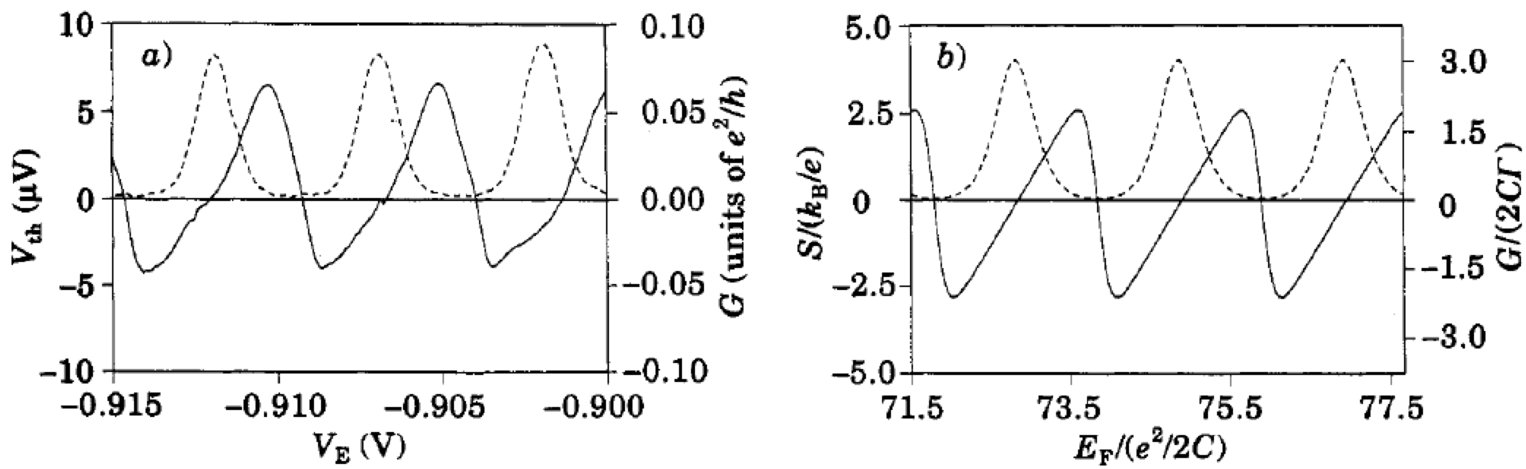


Thermopower of a QD

sequential tunneling



Large, metallic-like QD
N ~ 300
T ~ 230 mK
E_C ~ 0.3 meV
$E_C / k_B T$ ~ 15

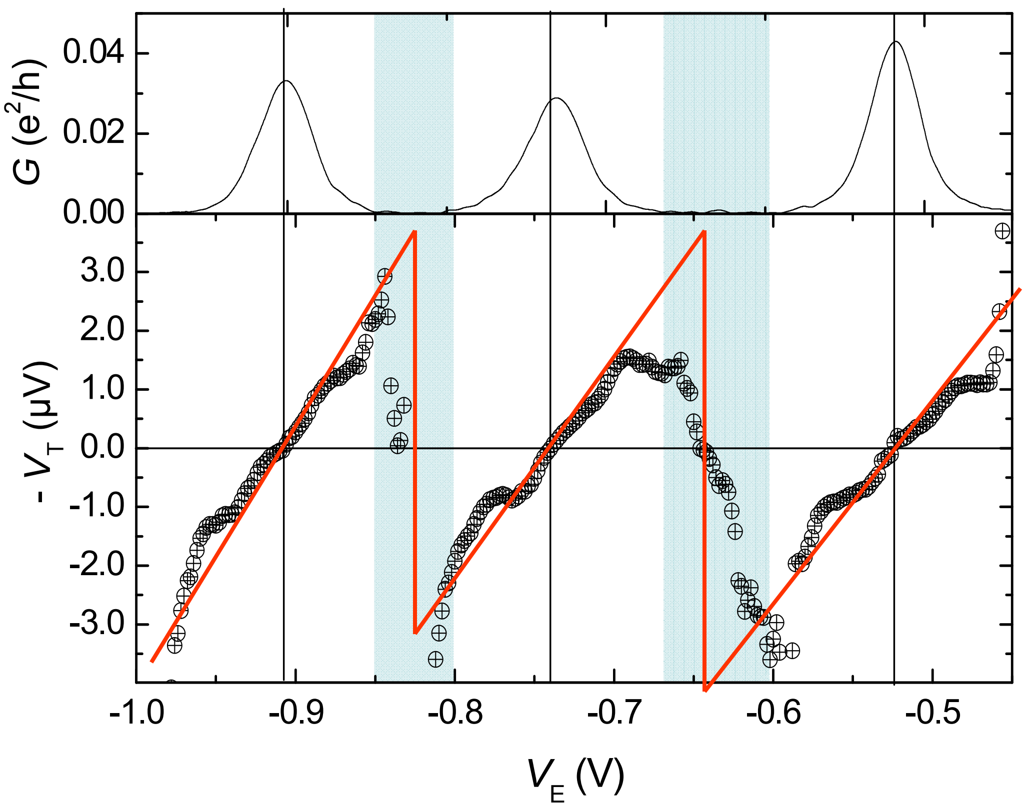


A.A.M. Staring et al., Europhys. Lett. 22, 57 (1993).

Thermopower of a QD

sequential tunneling

$\propto S$

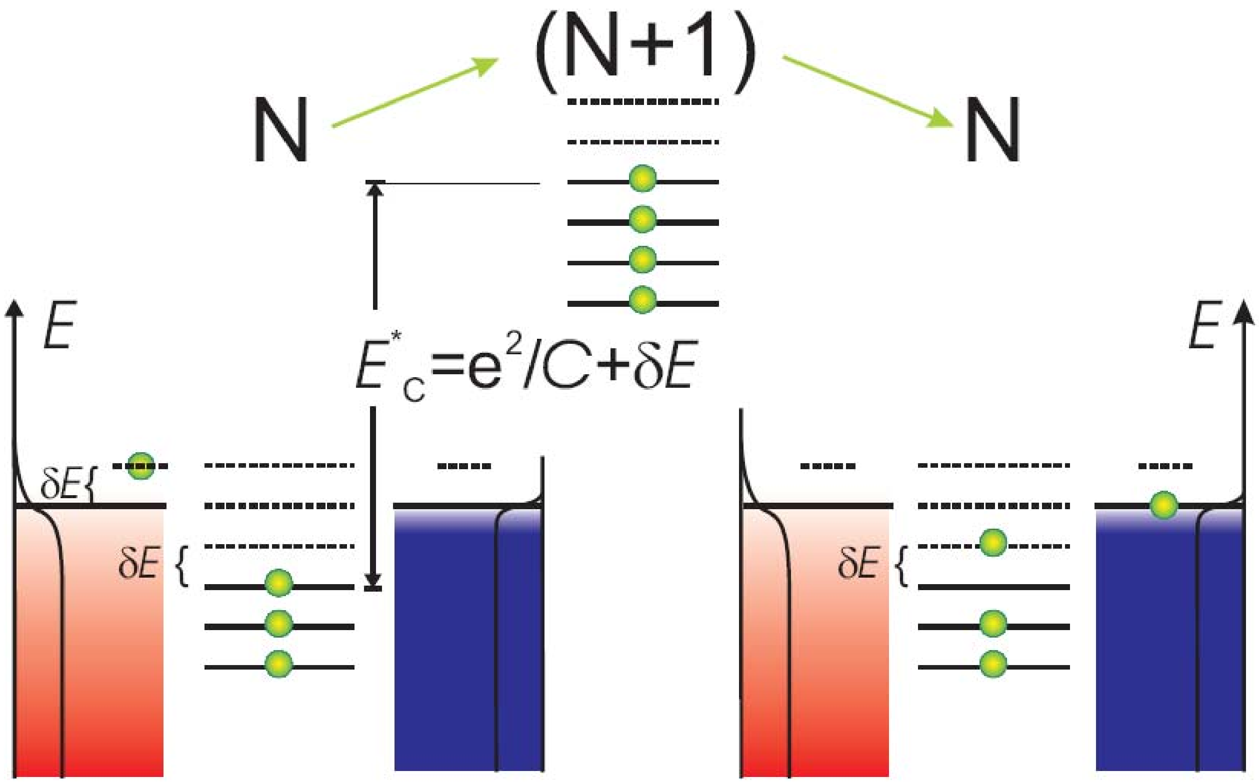


small QD
N ~ 15
T ~ 1.5 K
E_C ~ 2 meV
$E_C / k_B T$ ~ 15

Sample:
Bo_I13C

Thermopower of a QD

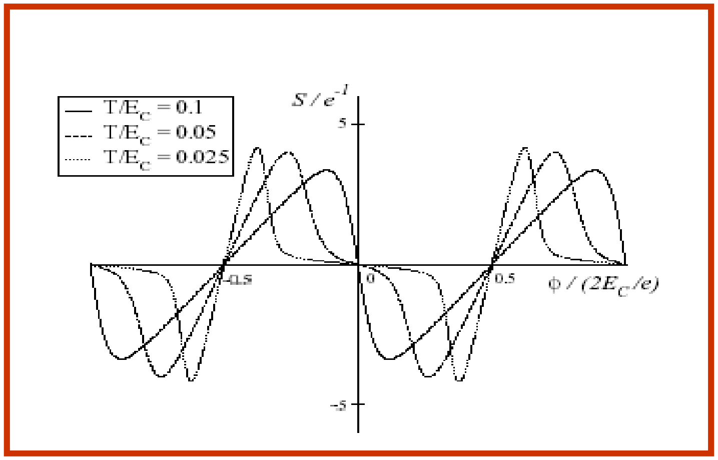
cotunneling contribution



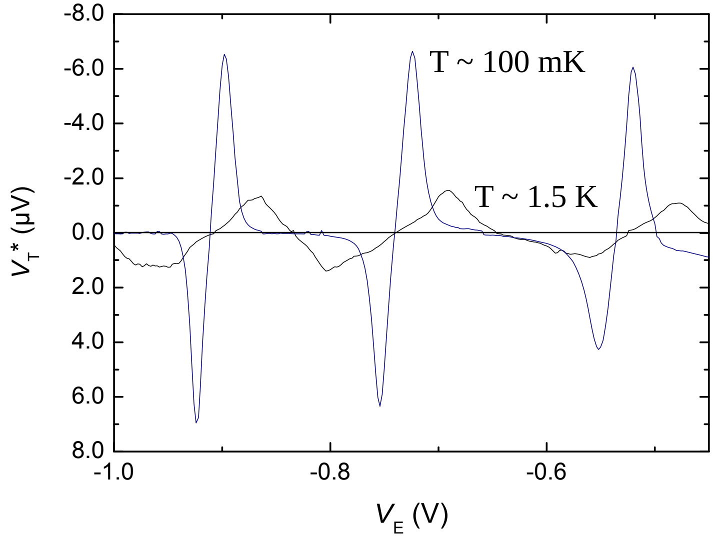
suppression of thermovoltage

Thermopower of a QD

cotunneling contribution



[M. Turek and K.A. Matveev, PRB, **65**, 115332 (2001)]



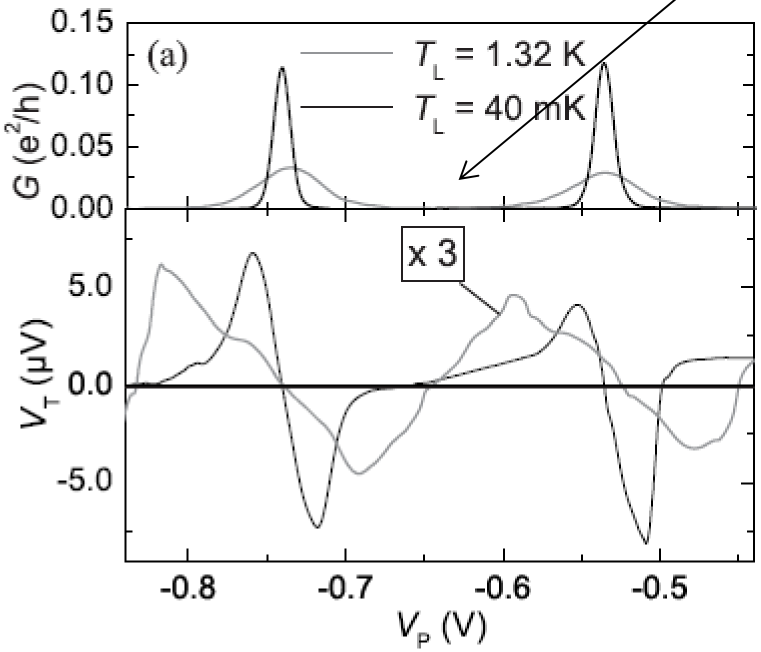
N ~ 15
E_C ~ 2 meV
$E_C / k_B T$ ~ 230

R. Scheibner et al., PRB **75**, 041301 (2007)

Thermopower of a QD

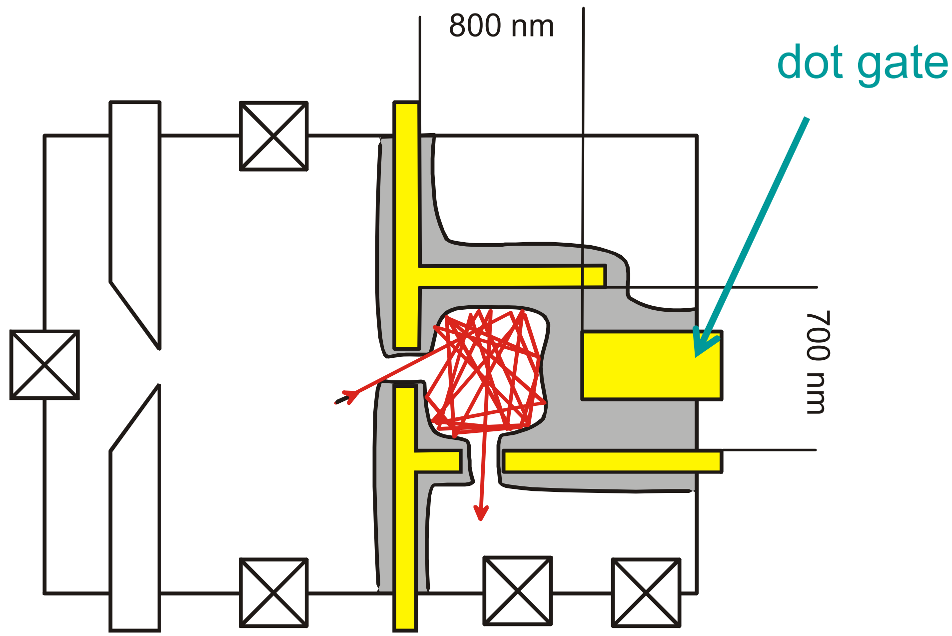
cotunneling contribution

no signatures of cotunneling processes in the CB regime



R. Scheibner et al., PRB **75**, 041301 (2007)

Chaotic Quantum Dot



n_s = 3.4 x 10^{11} cm^{-2}

μ = 1 x 10^6 cm^2 / (V sec)

$G_{qpc} = 4\, e^2 / h$

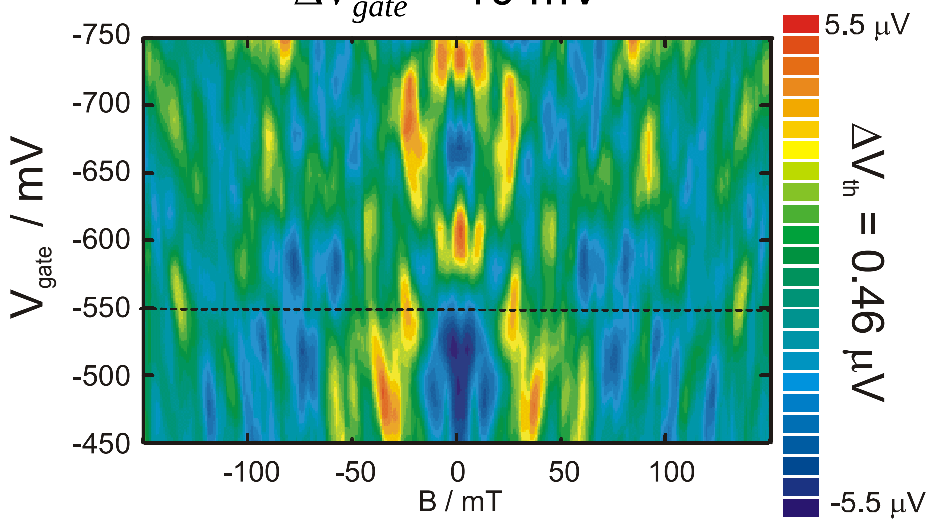
$(N_{qpc} = 2\,)$

Thermovoltage Fluctuations

T = 20 mK

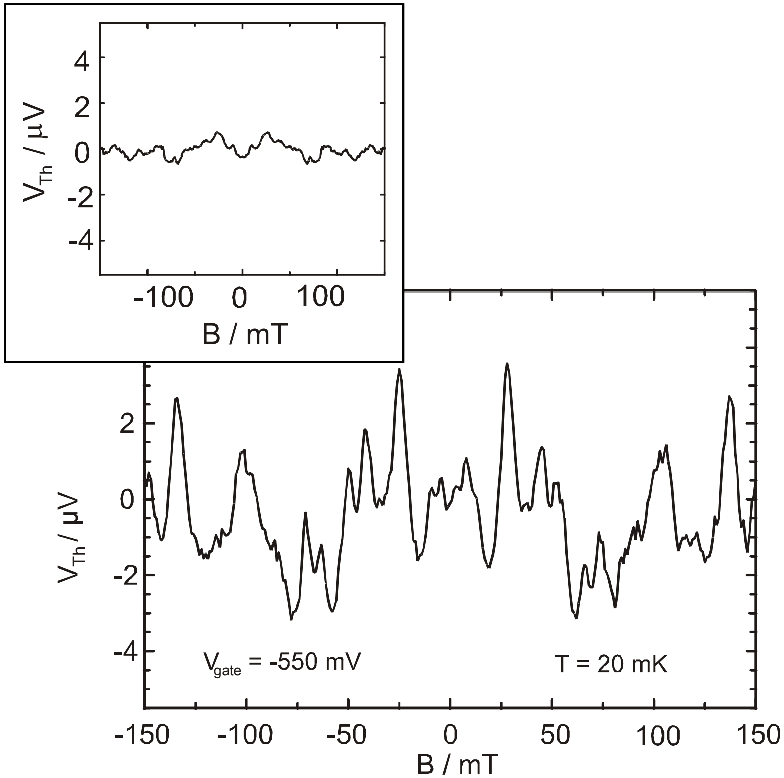
statistical ensemble

ΔV_{gate} = 10 mV



$I_{heating}$ = 40 nA

$\Delta T \approx$ 235 mK



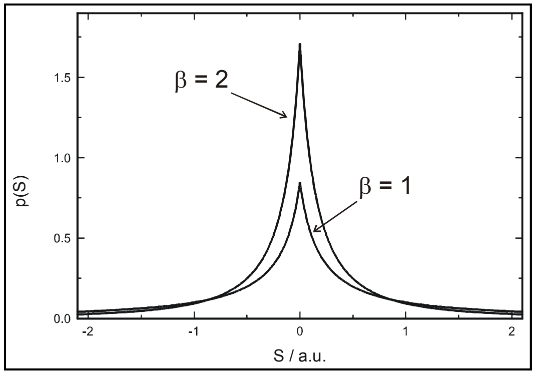
V_{gate} = - 550 mV

Thermopower Fluctuation Distribution

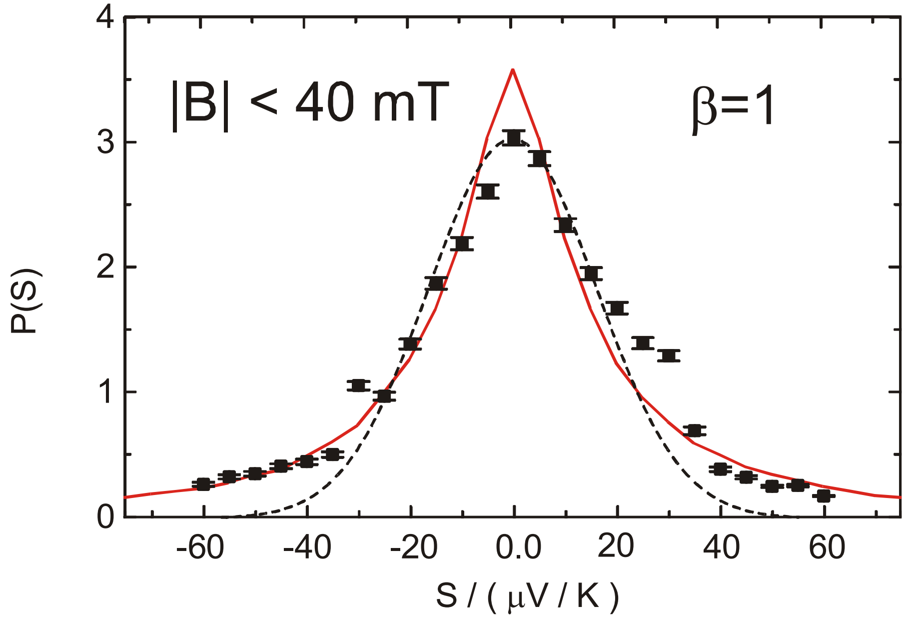
$$\frac{\partial G}{\partial E} = c(\tau_1 - \tau_2)\sqrt{G(1-G)}$$

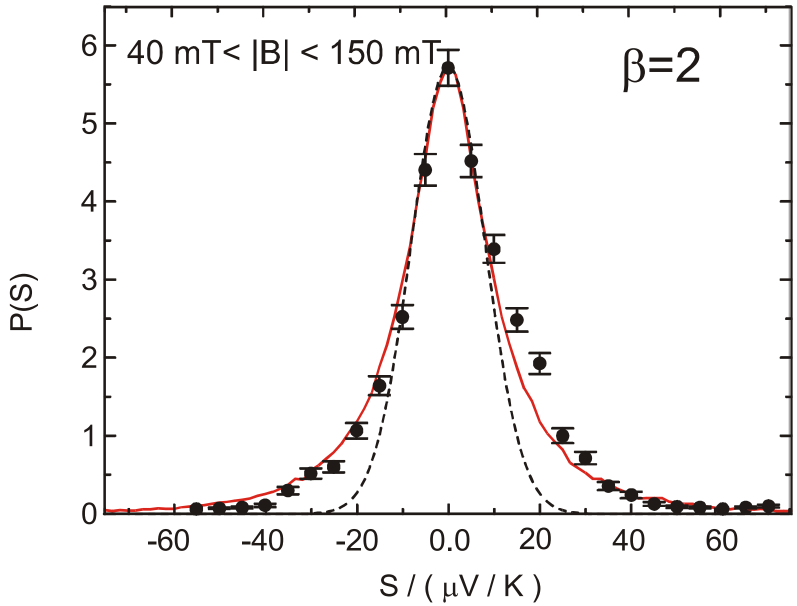
RMT:

analytic form for $N_1 = N_2 = 1$



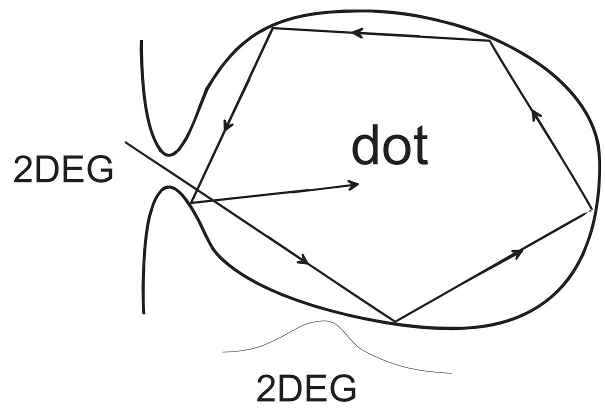
here: $N_1 = N_2 = 2$





S. Godijn et al., PRL **82**, 2927 (1999)

Residual Charging Energy



characteristic time scale:

$$\tau_{erg} = \Delta E / h$$

$$\tau_{dwell} = U^* / h$$

Luttinger liquid theory:

$$U^* = U_0 (1-t)^N$$

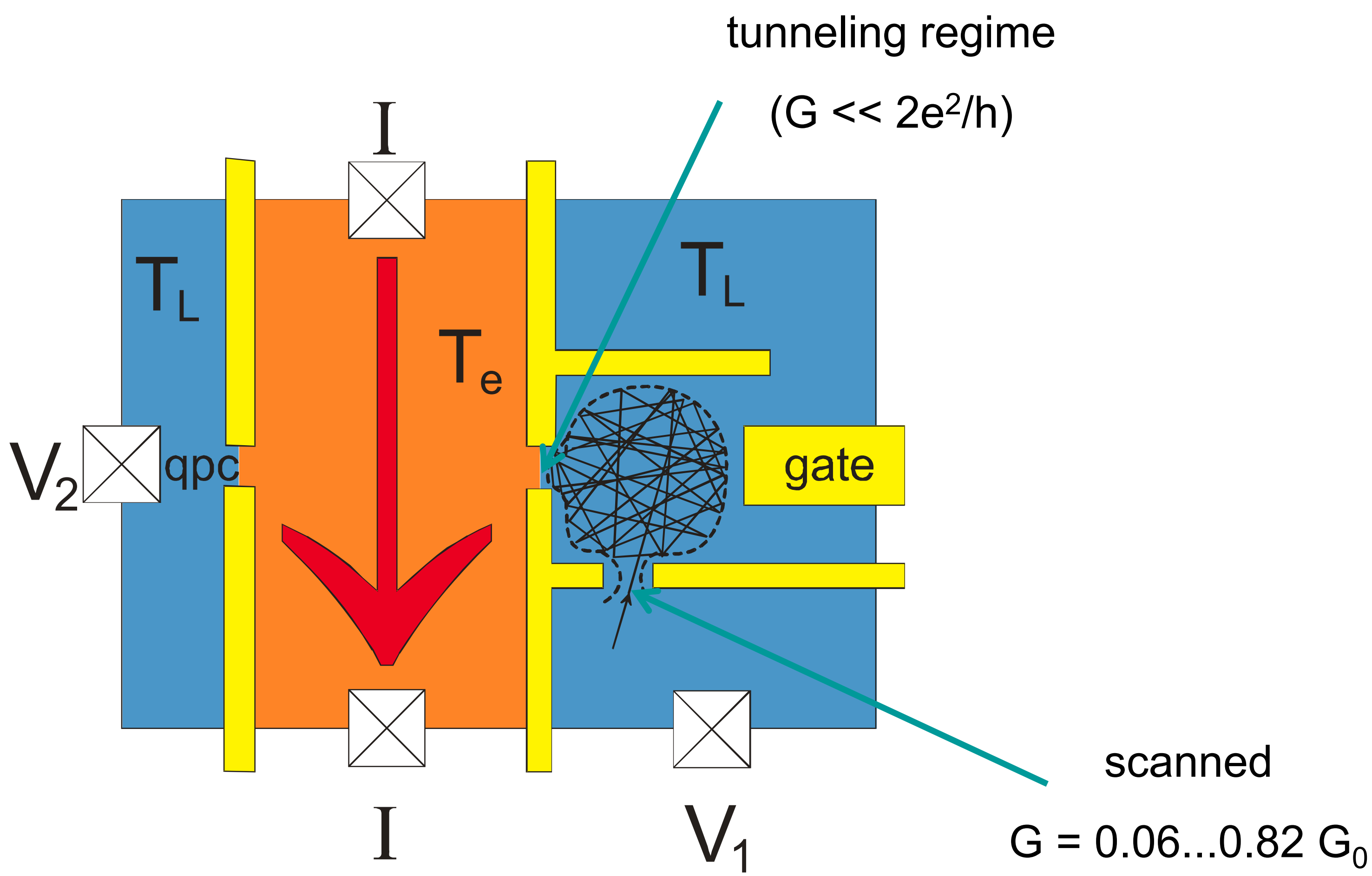
(Flensberg, 1993, 1994)

chaotic QD:

(Aleiner and Glazman, 1998)

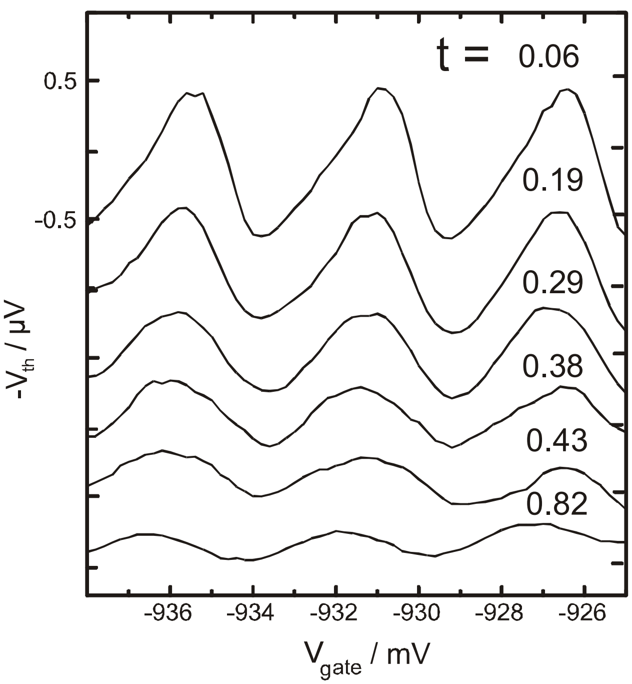
$$t \to 1 \Rightarrow \quad (1-t) \to \frac{\Delta E}{U_0} \ln^2\left(\frac{U_0}{\Delta E}\right)$$

Scaling Experiment

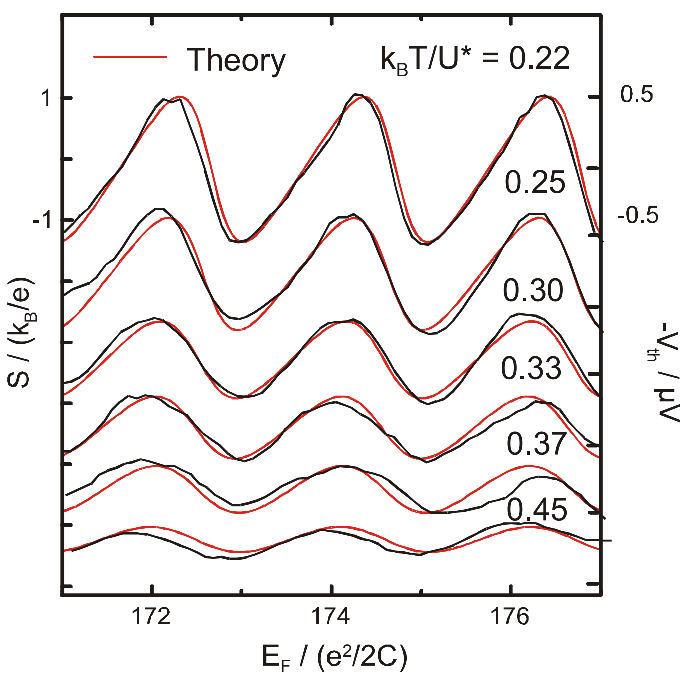


Scaling Results

Thermovoltage



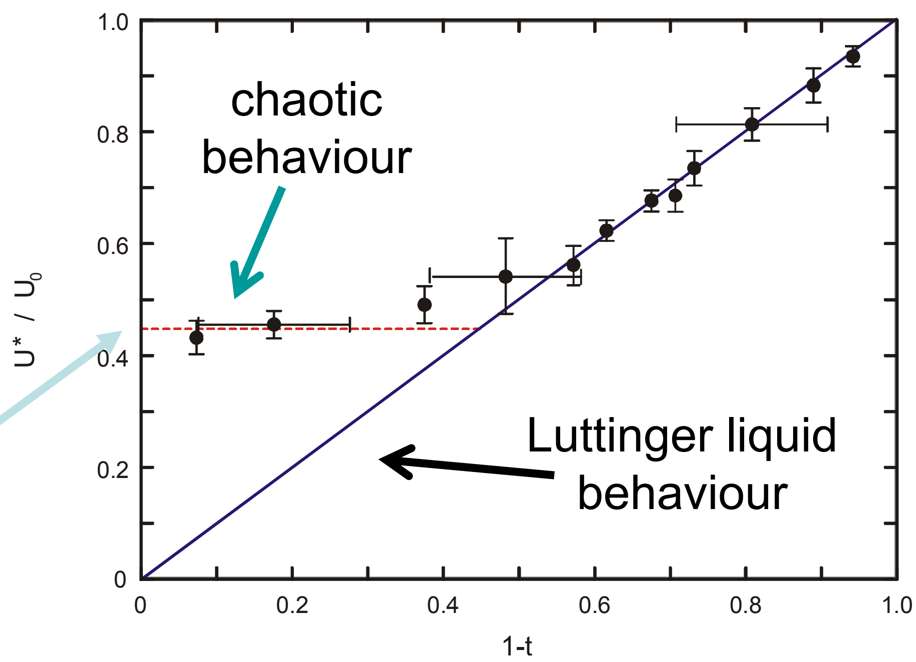
Thermopower



$I_{heating}$ = 40 nA

T_e = 255 mK, T_L = 40 mK

Scaling



extrapolation

$t \to 1$

$$U^*_{\exp}(t \to 1) \approx 0.45 U_0$$

theory

$\Delta E = 23\ \mu\text{eV}$

$U_0 = 100\ \mu\text{eV}$

$$U^* = U_0 \frac{\Delta E}{U_0} \ln^2\left(\frac{U_0}{\Delta E}\right) = 0.49 U_0$$

S. Möller et al., PRL **81**, 5197 (1998)

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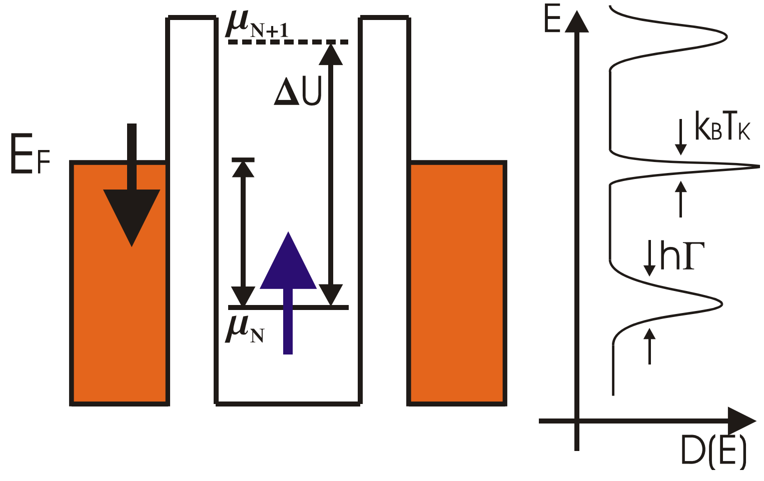
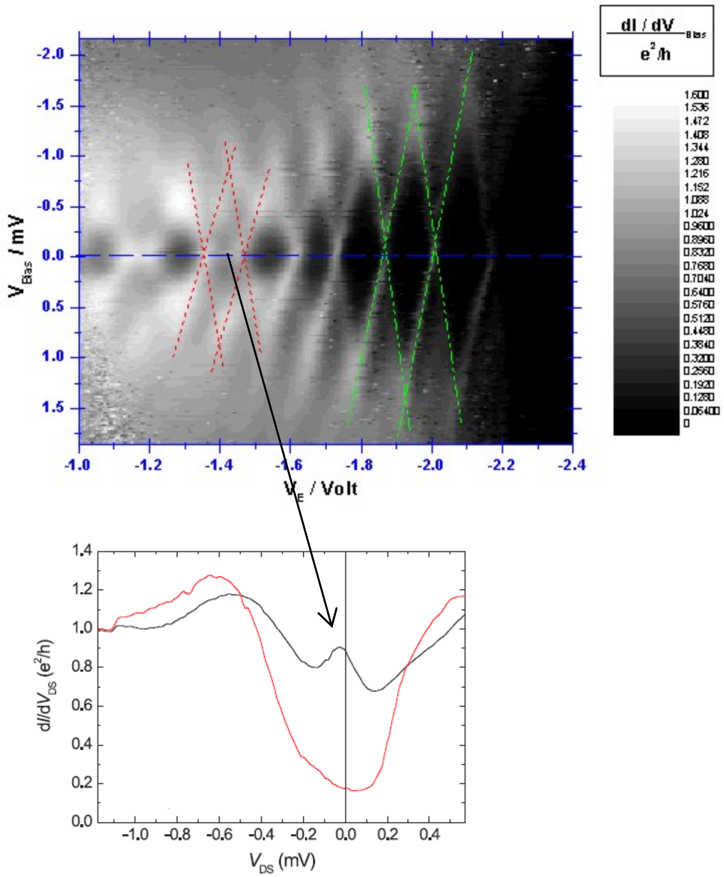
Spin-Correlated QD

EP 3

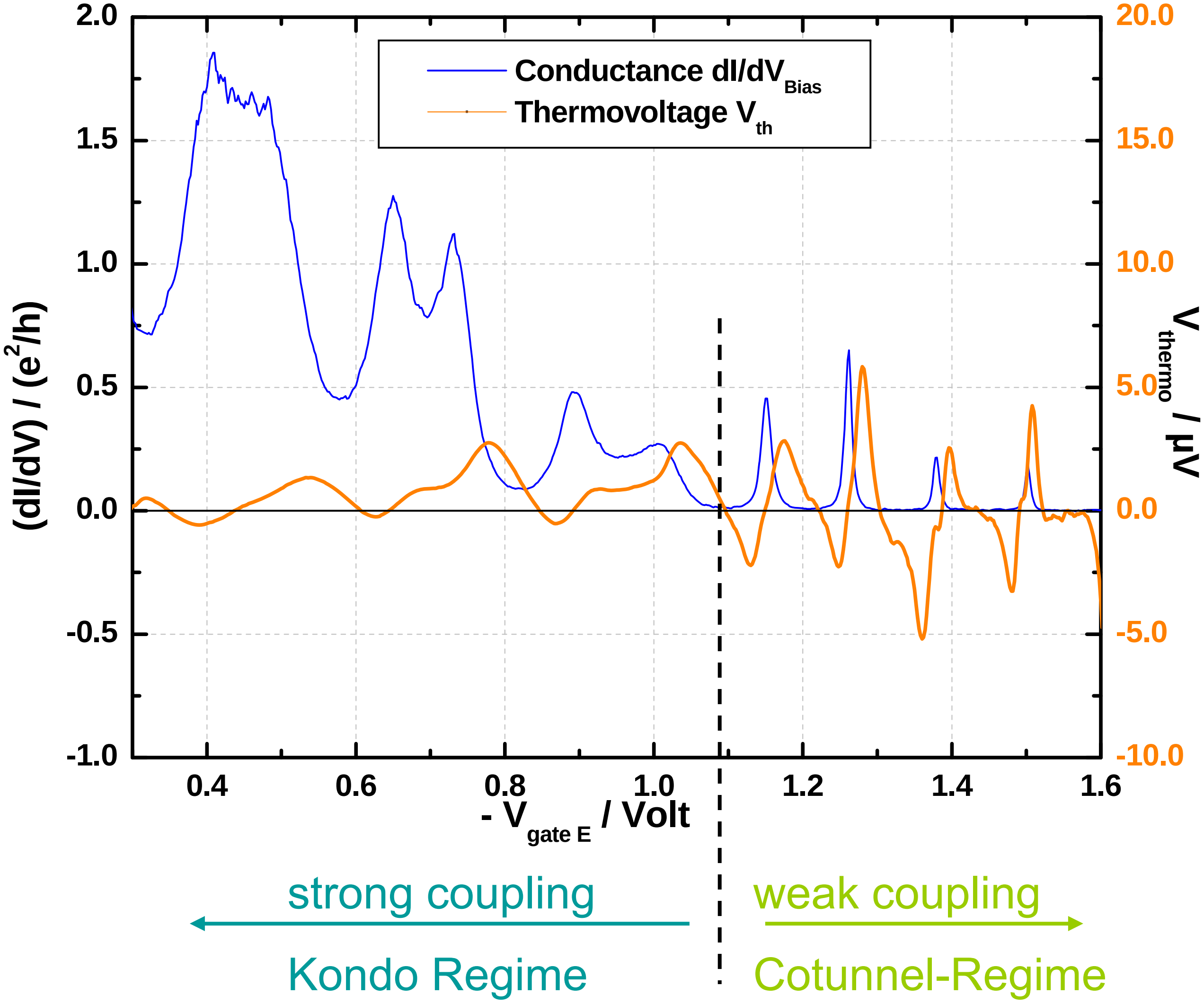
- existence of a magnetic moment on the QD can lift the CB
- transport mechanism: spin scattering
- hybridization of free electrons in the leads with localized magnetic moment leads to resonance at the Fermi edge

Kondo Resonance

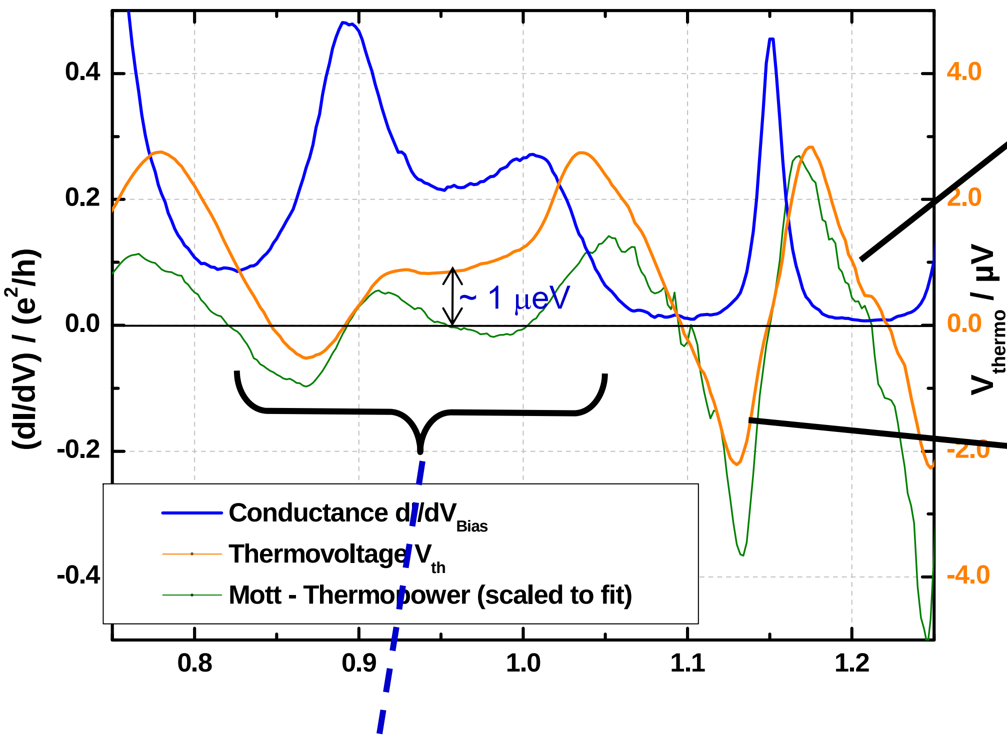


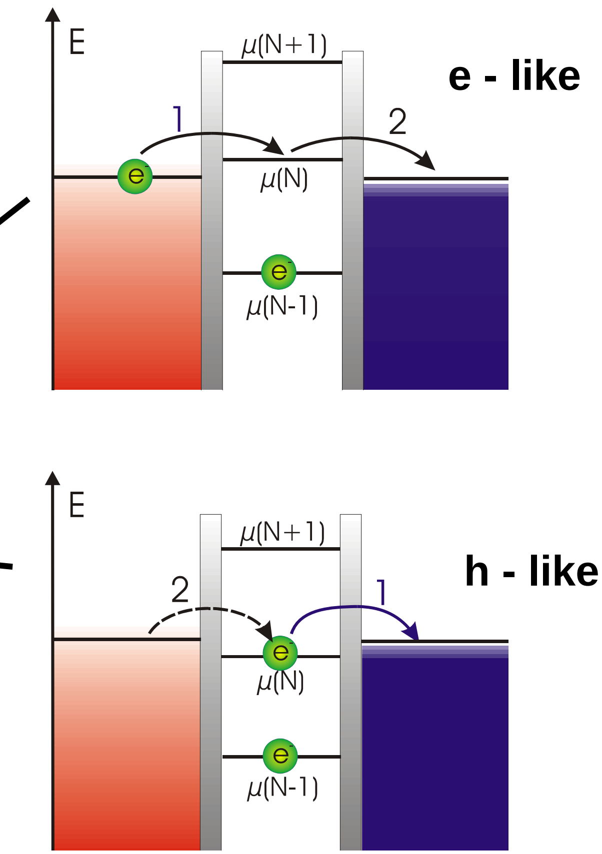


Spin-Correlated QD



Spin-Correlated QD





Asymmetry between
electron- and hole-like transport:
Mixed-valence regime

Scheibner *et al*. PRL **95**, 176602 (2005)

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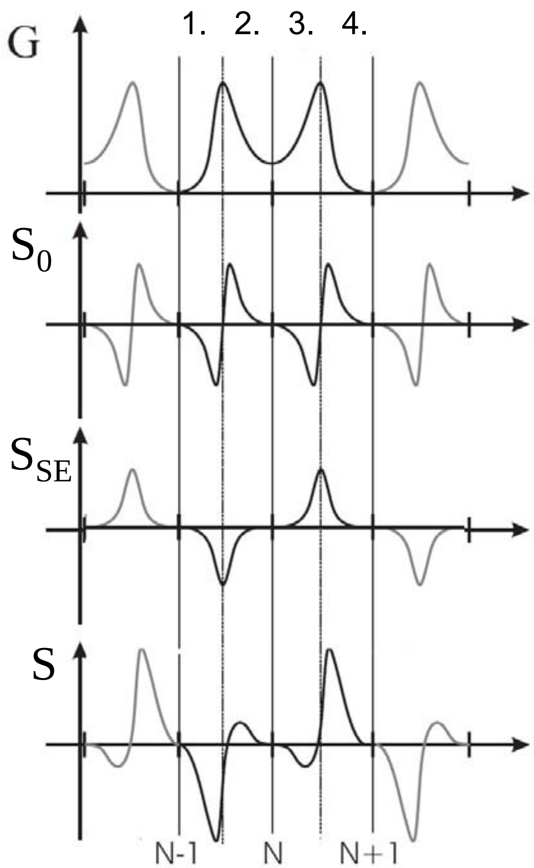
Spin-Entropy Transport

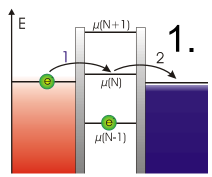
EP 3

Entropy change ΔS
adding one electron to an empty site: $\Delta S = k_B(\ln g_f - \ln g_i) = k_B(\ln 2 - \ln 1) = k_B \ln 2$

$-V_g$

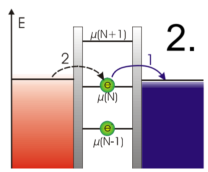


$g_s = 1$ $g_s = 2$ $g_s = 1$



1. e-like transport from the hot to the cold reservoir
$\Delta S = k_B(\ln g_f - \ln g_i) = k_B(\ln 2 - \ln 1) = k_B \ln 2$

➔ $S_{SE}=-k_B/e \ln 2$



2. h-like transport from the cold to the hot reservoir
$\Delta S = k_B(\ln g_f - \ln g_i) = k_B(\ln 1 - \ln 2) = -k_B \ln 2$

➔ $S_{SE}=-k_B/e \ln 2$

3. e-like transport from the hot to the cold reservoir
$\Delta S = k_B(\ln g_f - \ln g_i) = k_B(\ln 1 - \ln 2) = -k_B \ln 2$

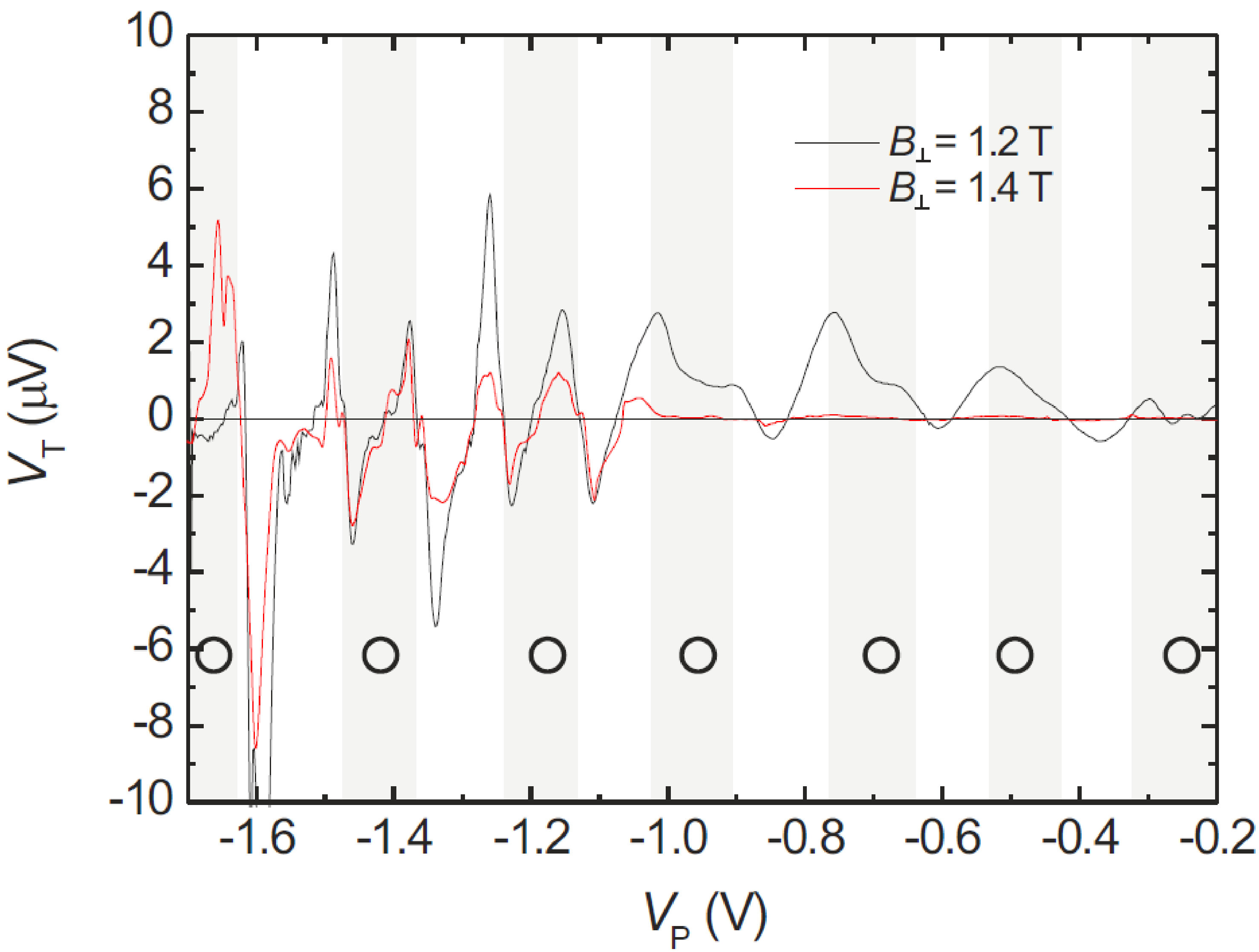
➔ $S_{SE}=k_B/e \ln 2$

4. h-like transport from the cold to the hot reservoir
$\Delta S = k_B(\ln g_f - \ln g_i) = k_B(\ln 2 - \ln 1) = k_B \ln 2$

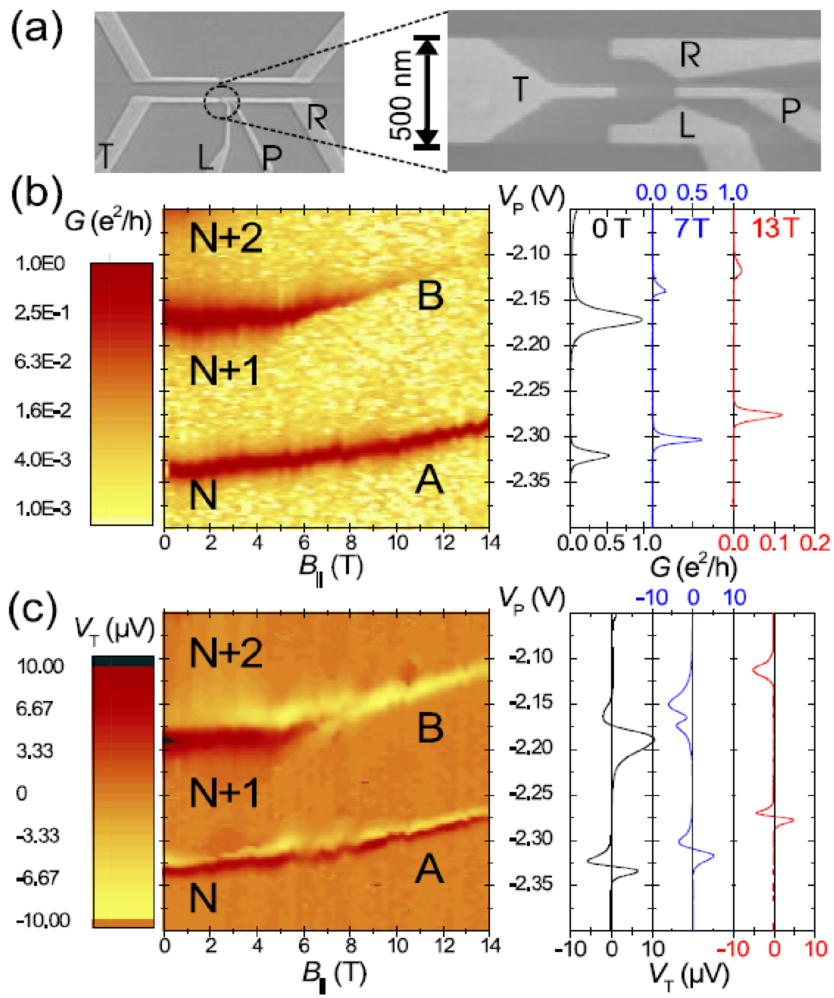
➔ $S_{SE}=k_B/e \ln 2$

R. Scheibner et al., PRL **95**, 176602 (2005)
R. Scheibner, PhD-Thesis, Würzburg 2007

Spin entropy contributions

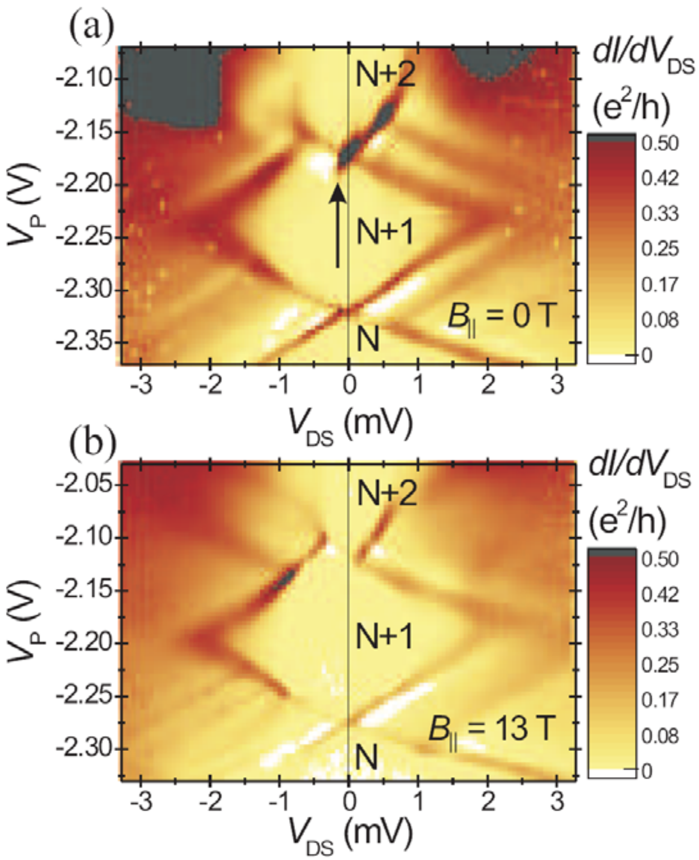


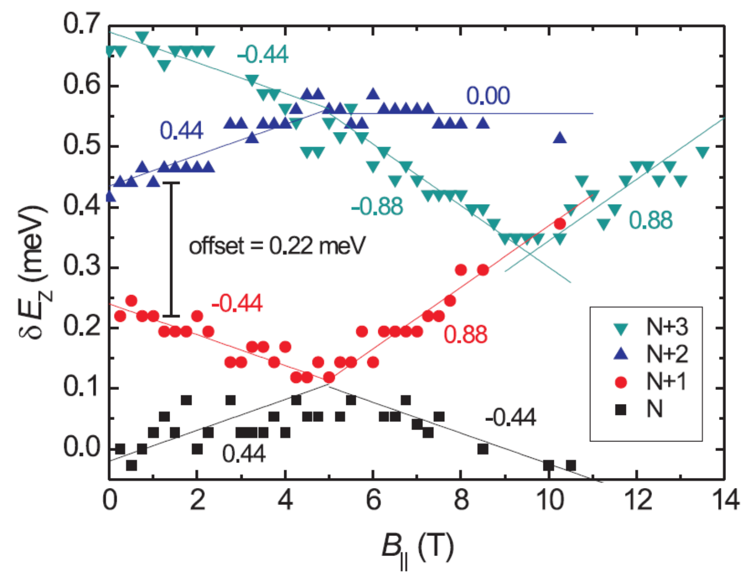
Thermal Rectifier



R. Scheibner et al., NJP **10**, 083016 (2008)

Thermal Rectifier

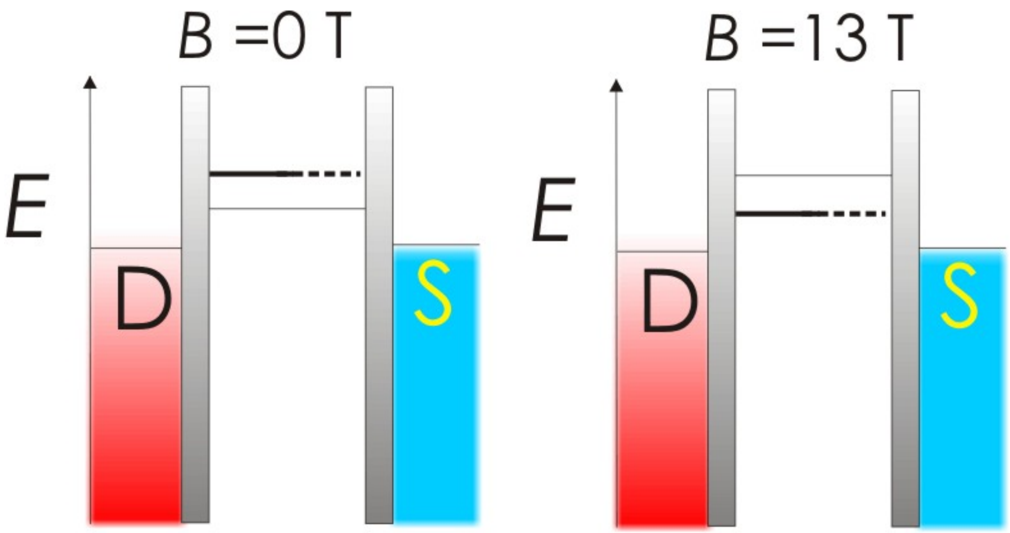


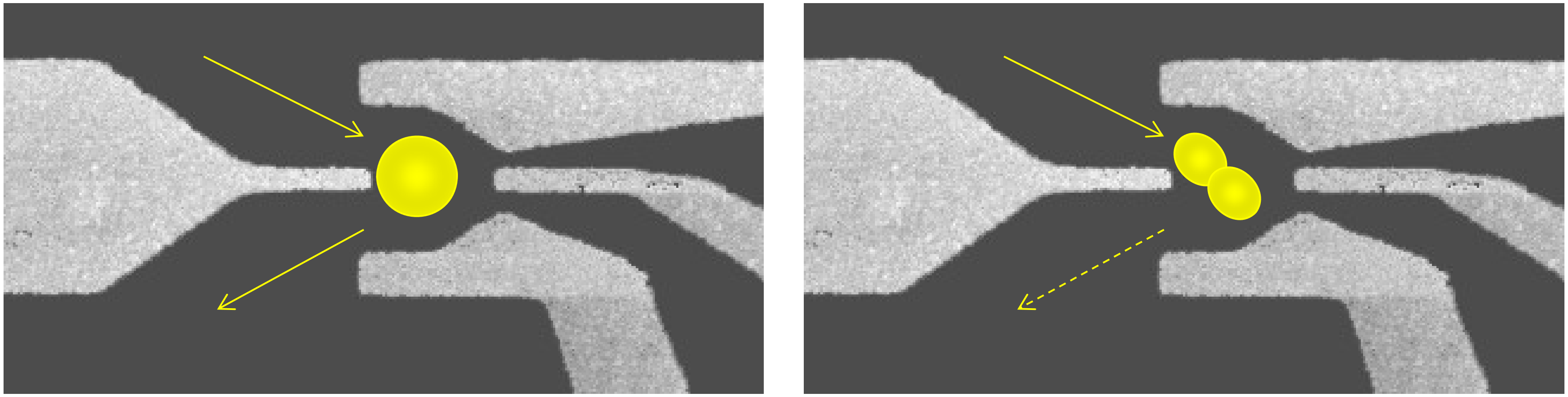


R. Scheibner et al., NJP **10**, 083016 (2008)

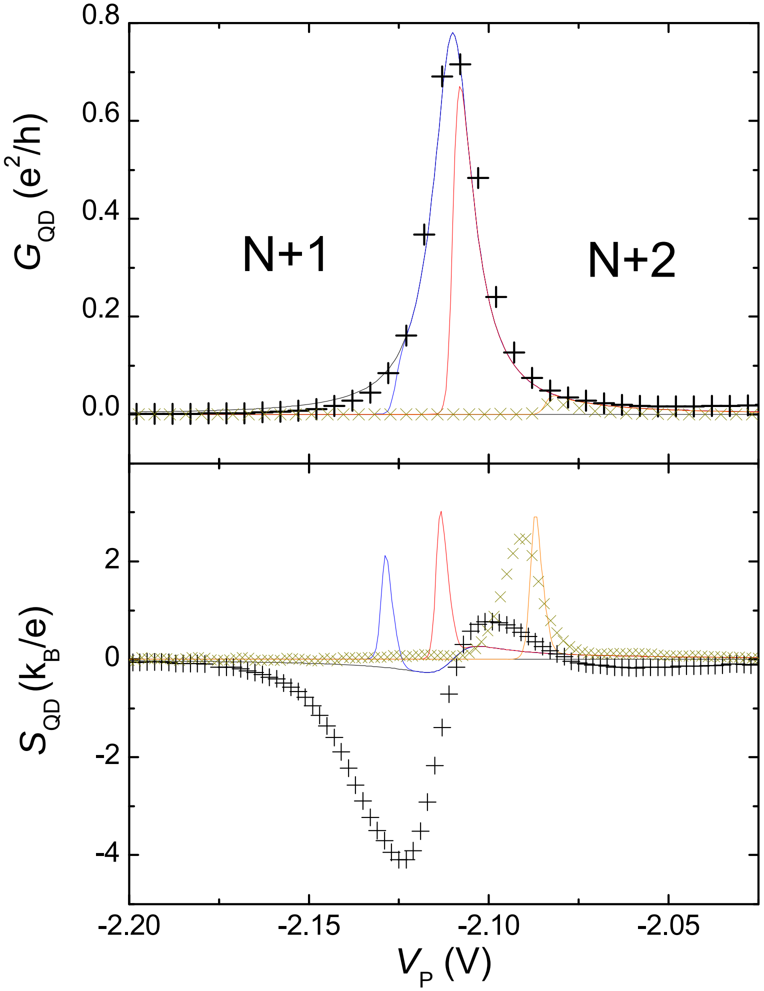
Thermal Rectifier

asymmetrically coupled states





Thermal Rectifier



$$J_{tot} = \int_{-\infty}^{\infty} dE \left(\frac{\Lambda}{h}\right)\left[f_L(E,T) - f_R(E,T)\right]t(E)$$

$$\Lambda = \begin{cases} -e & \text{(electron)} \\ E-\mu & \text{(energy)} \end{cases}$$

$$S = -\frac{L_{12}}{L_{11}} = -\frac{\left(-\frac{e}{Th}\right)\int_{-\infty}^{\infty} dE\,(E-\mu)t(E)\left(-\frac{df}{dE}\right)}{\left(\frac{e^2}{h}\right)\int_{-\infty}^{\infty} dE\,t(E)\left(-\frac{df}{dE}\right)}$$

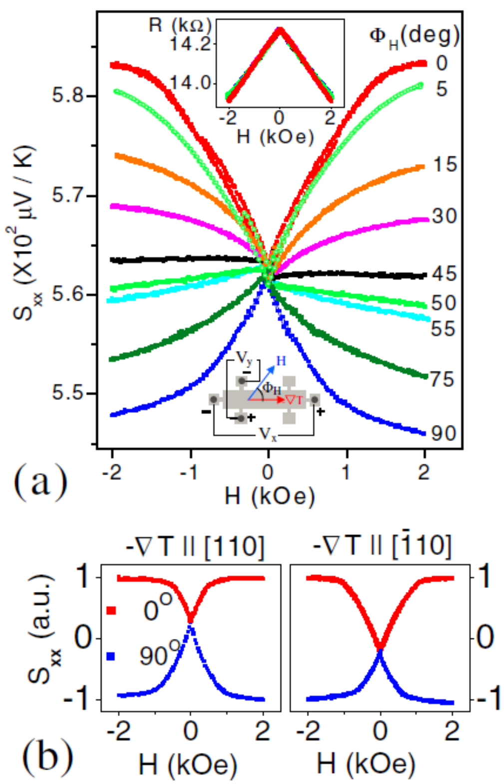
$$t(E) = A\frac{(\Gamma/2)^2}{(\Gamma/2)^2 + E^2} \times f(E - \delta E_Z, T)$$

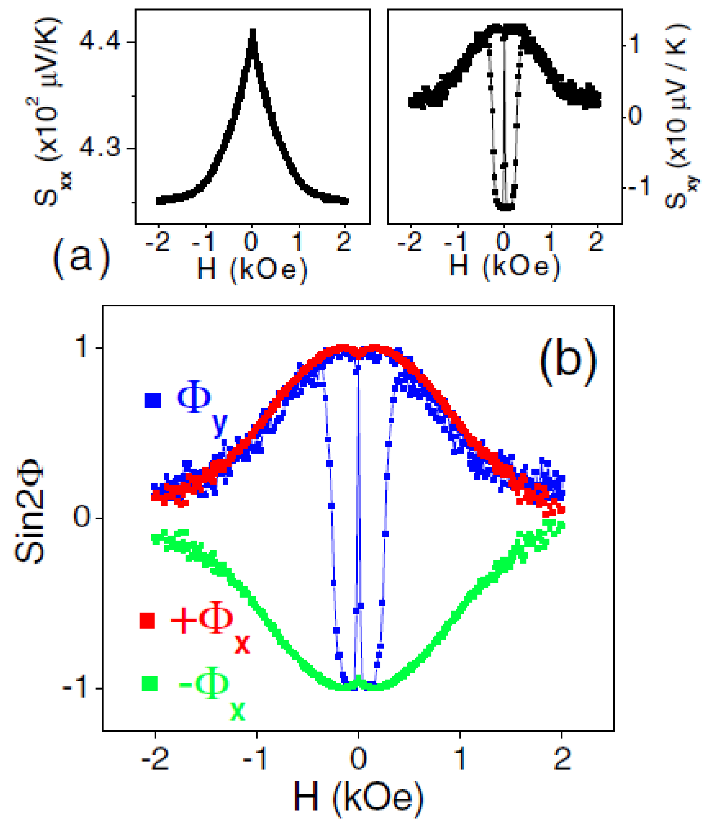
R. Scheibner et al., NJP **10**, 083016 (2008)

Thermopower of a (Ga,Mn)As based M-I-FM junction

Thermopower in (Ga,Mn)As

First (Ga,Mn)As data by Shi group (Pu et al., Phys. Rev. Lett. **97**, 036601 (2006).



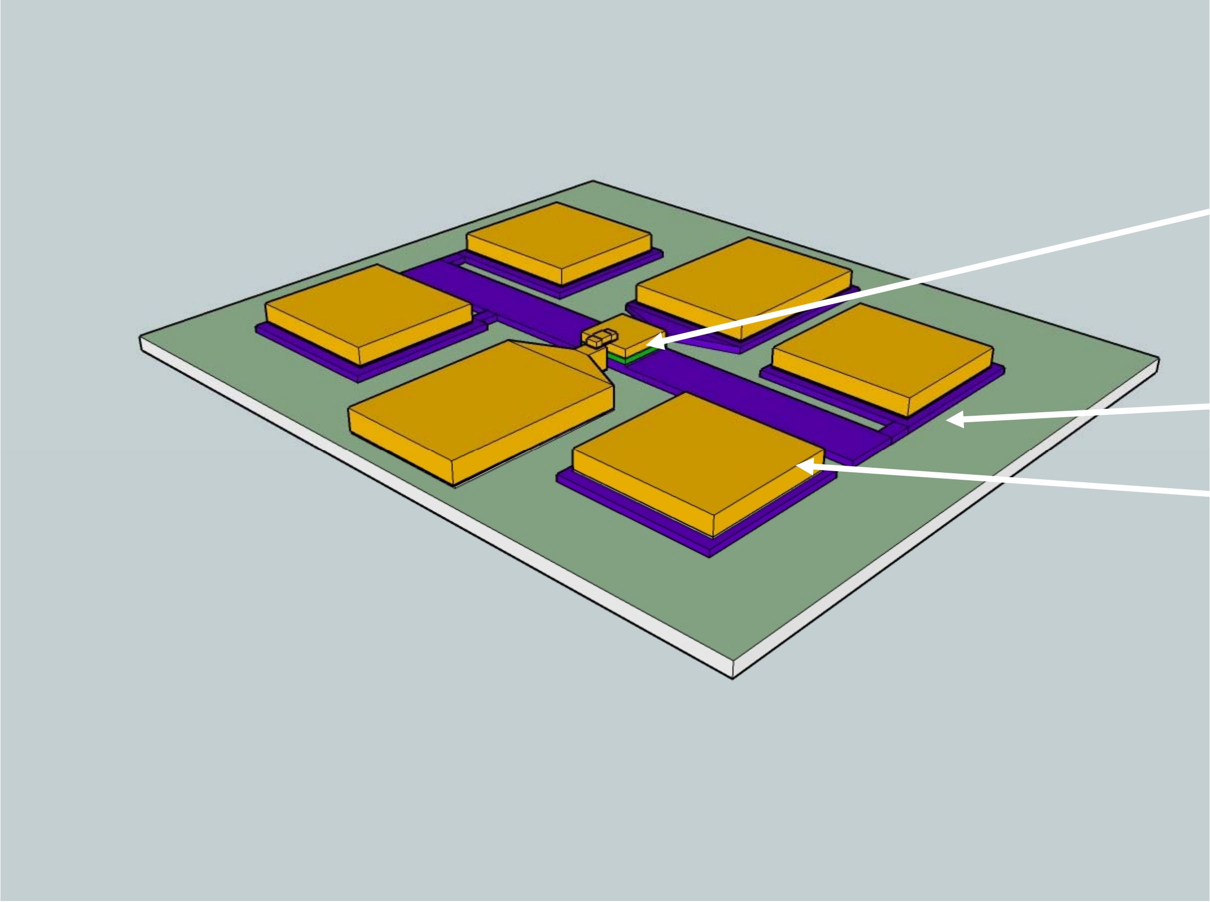


- Signal too large compared to band model
  (claim Fermi level in impurity band)
- Dependence on field direction mimics AMR, not band structure

Data dominated by phonon drag?

Measure Diffusion Thermopower, only.

Apply current heating technique to (Ga,Mn)As



20 nm (Ga,Mn)As (3% Mn)

60 nm n-GaAs (2×10^{19} cm^{-3})

ohmics 5nm Ti/ 30 nm Au

No counter point contact: T_{el} inferred from weak localization peak in channel


30nm Au
5nm Ti
20nm LT GaMnAs
1nm LT GaAs
60nm n+- GaAs
200nm HT GaAs
GaAs

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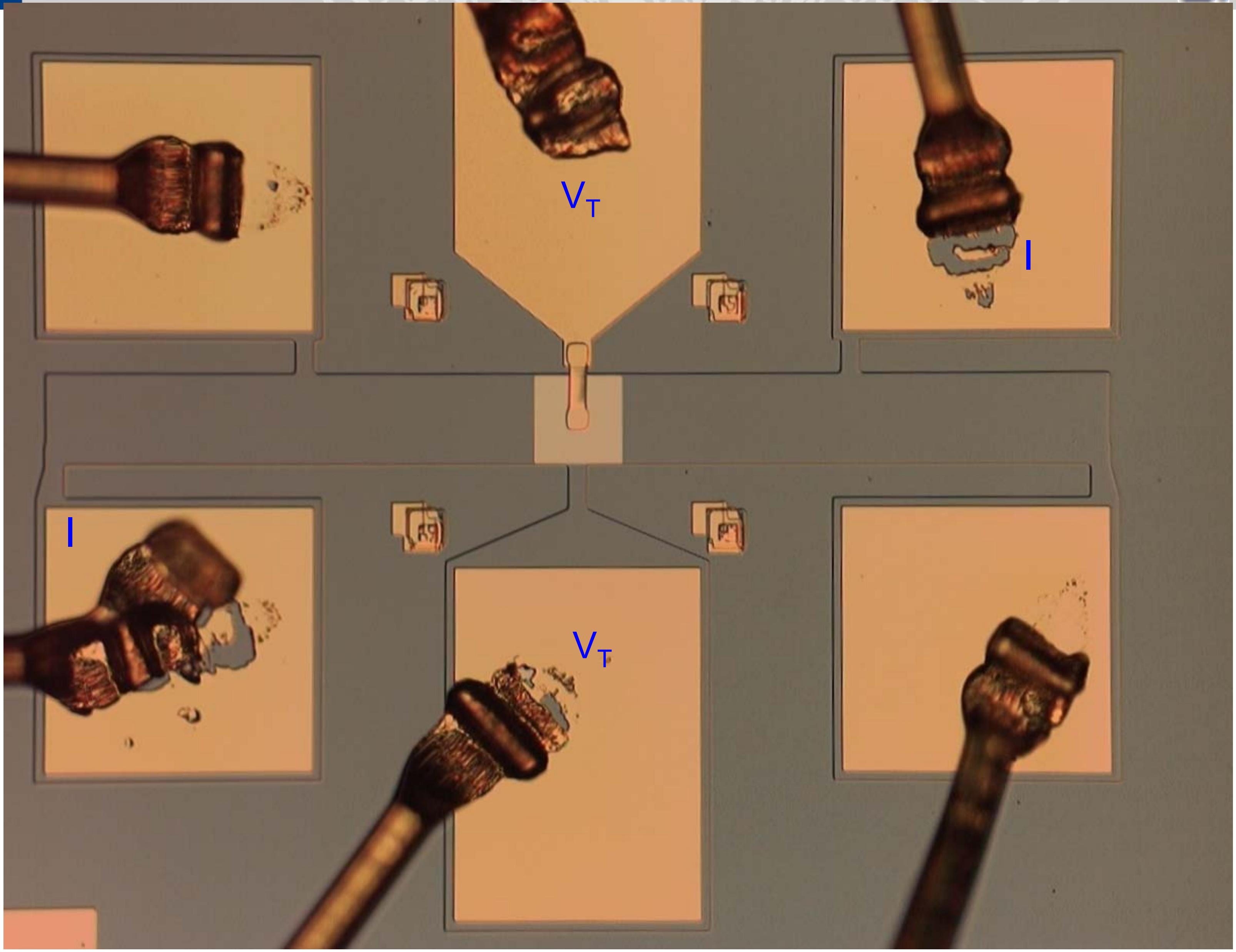
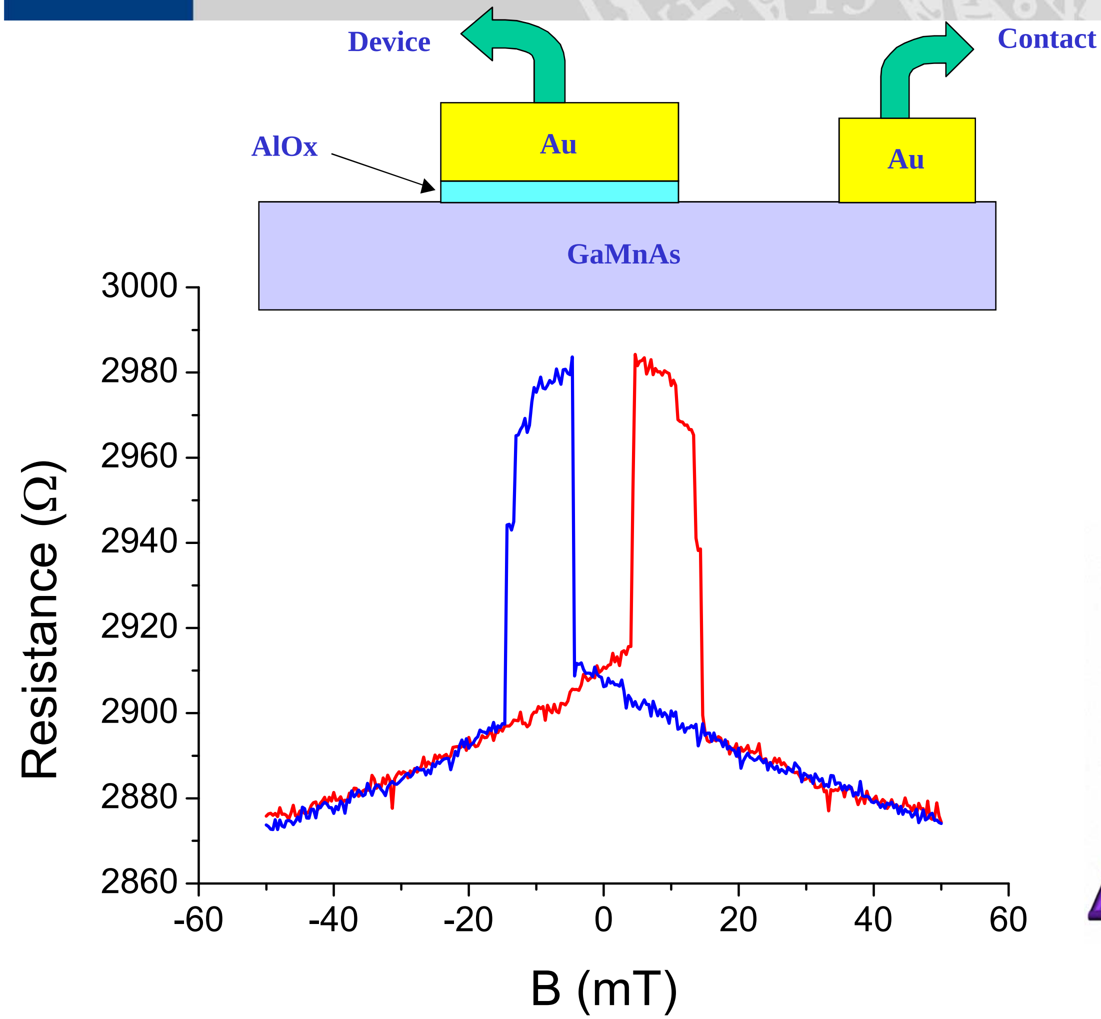
Large Device (no UTF)

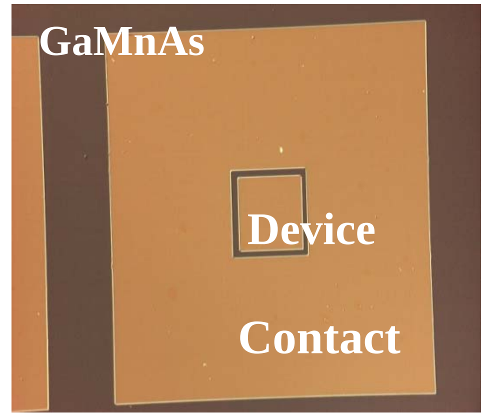
EP 3

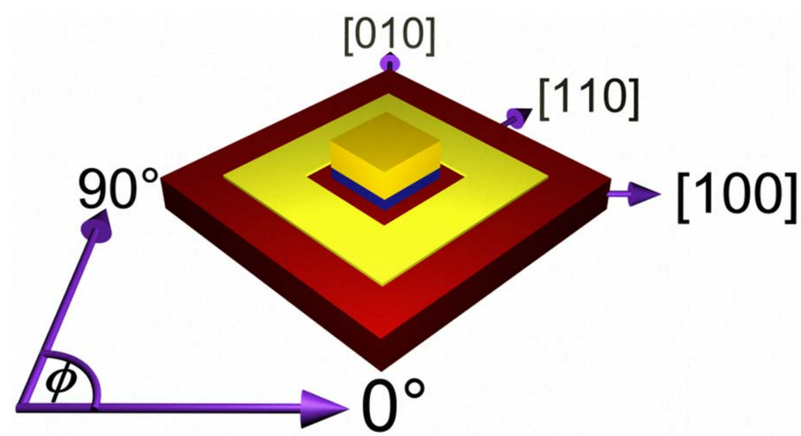


Flashback: TAMR in (Ga,Mn)As

C. Gould et al., Phys. Rev. Lett. **93**, 117203 (2004).

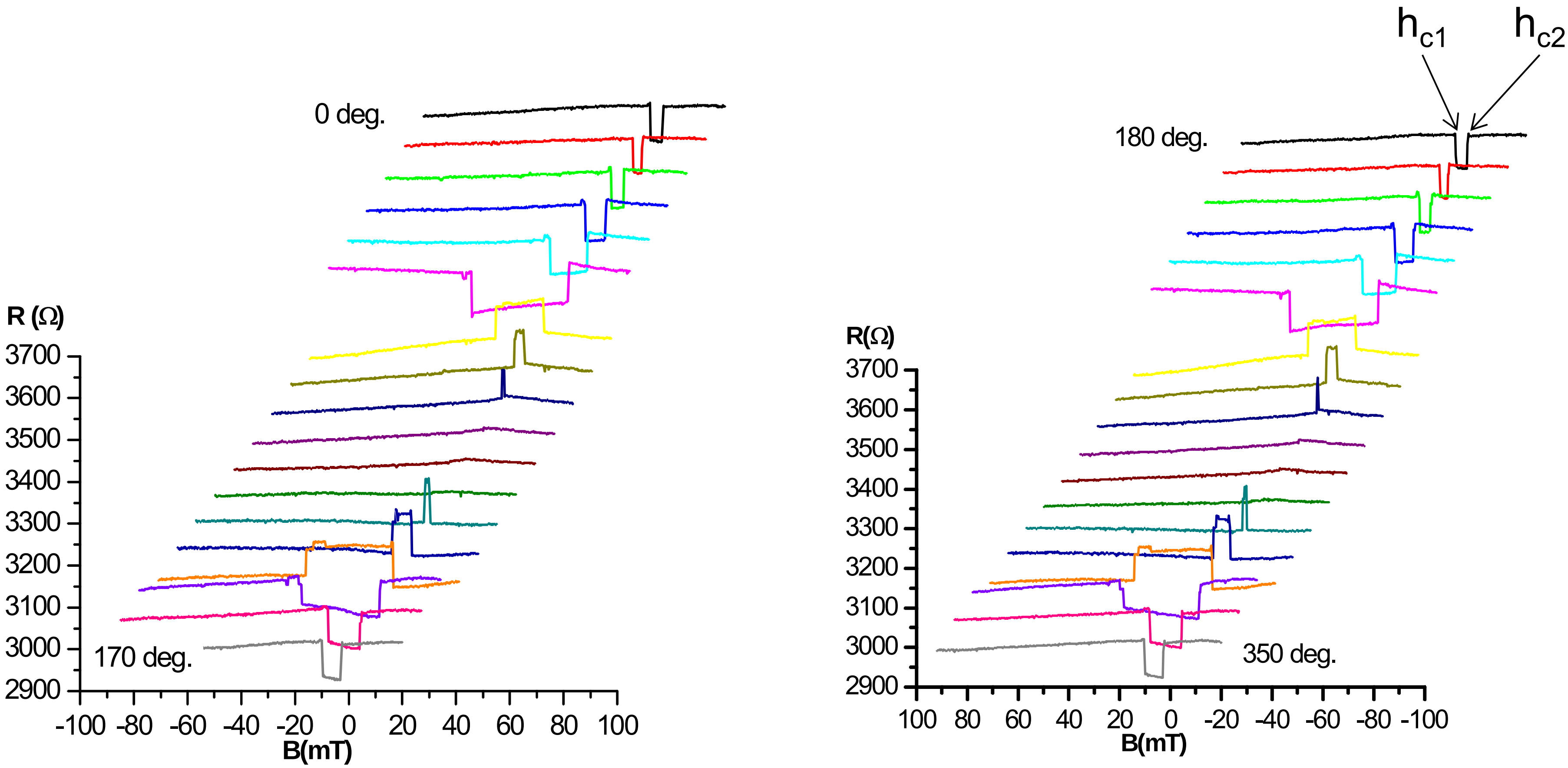






A tunnel barrier between a non-magnetic metal (Au) and ferromagnetic (Ga,Mn)As can exhibit a huge magnetoresistance that can show the signature of a spin valve.

Spin-Valve like TMR in (Ga,Mn)As/AlOx/Non-magnet devices

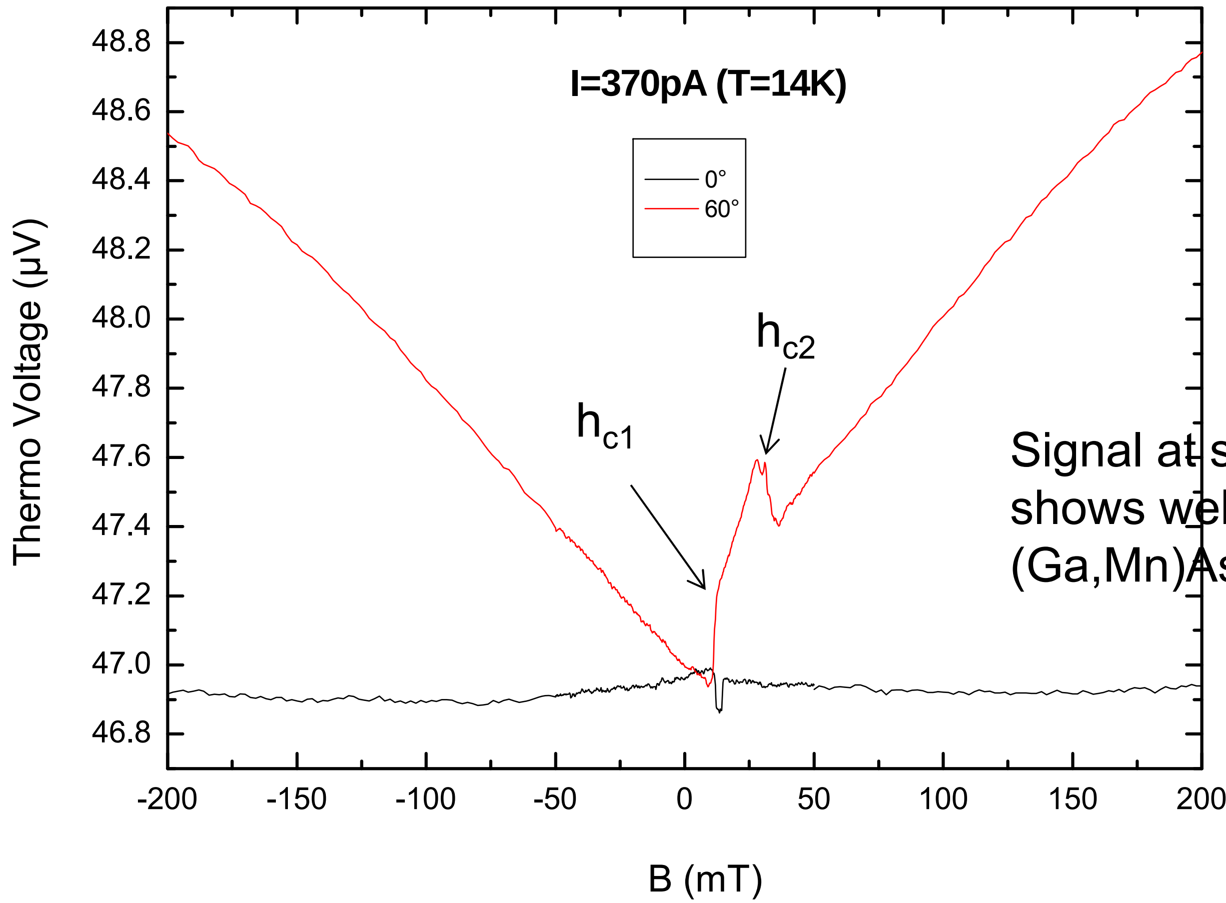


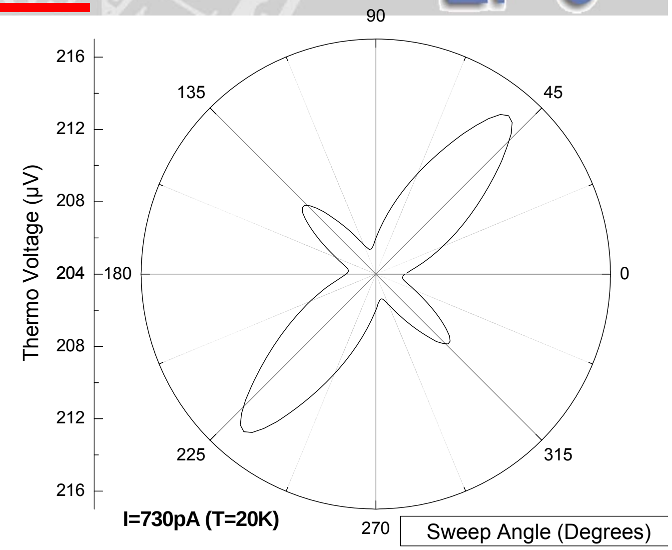
Dependence of the magnetoresistance effect on the in-plane field angle (angle with respect to [100]). Effect due to biaxial anisotropy and anisotropic d.o.s.

Now back to thermopower experiment....

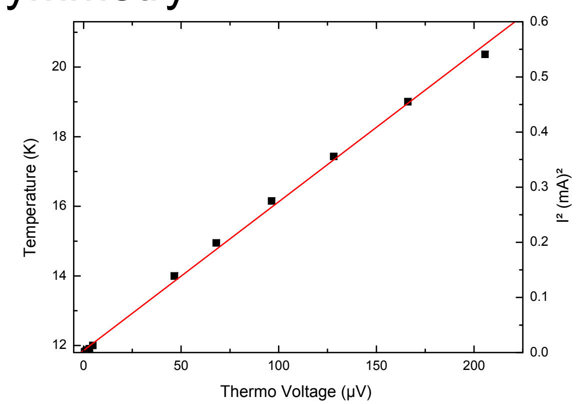
Tunnel Anisotropic Magneto Thermopower

T. Naydenova et al., Phys. Rev. Lett. 107, 197201 (2011)





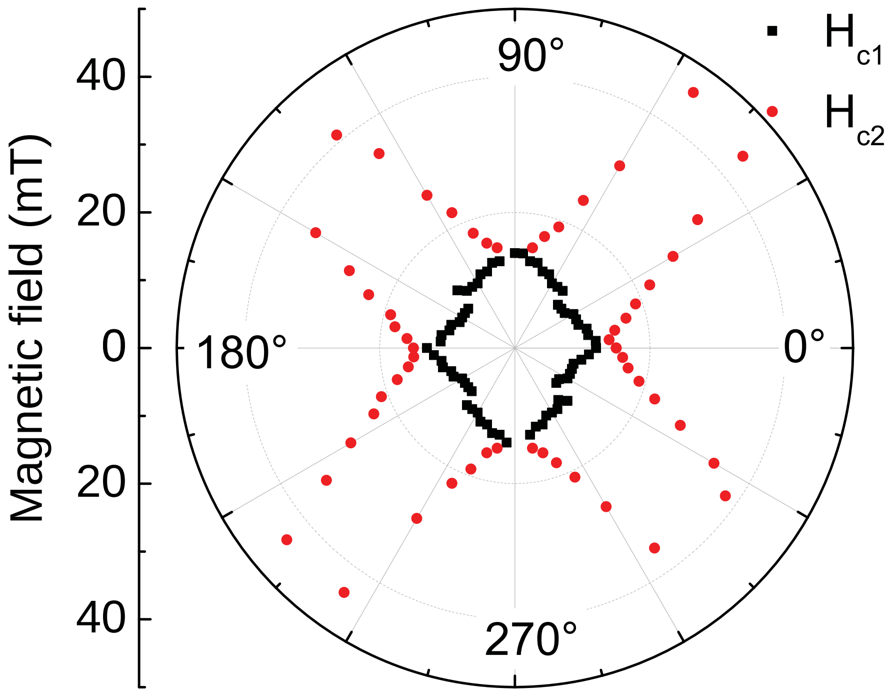
Signal at saturation (300 mT), shows well-known (Ga,Mn)As symmetry...

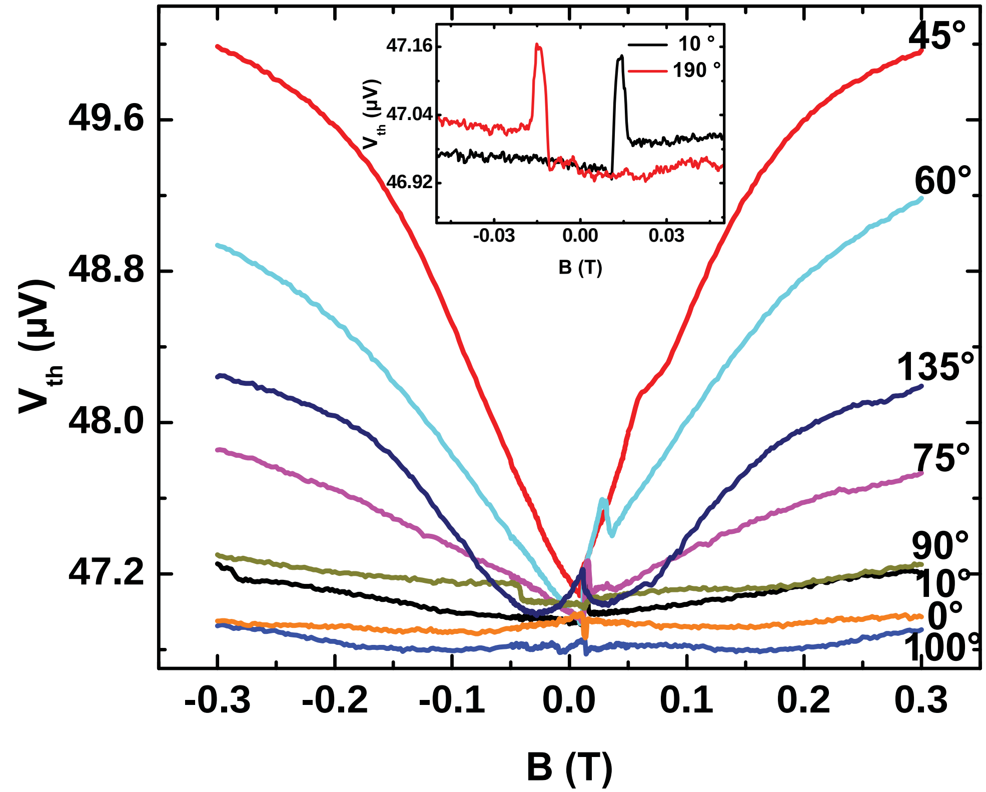


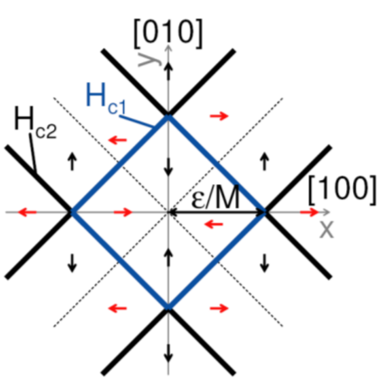
Signal with hysteresis, shows expected angle dependence

...and is quadratic with current

hc1 and hc2 for various field directions





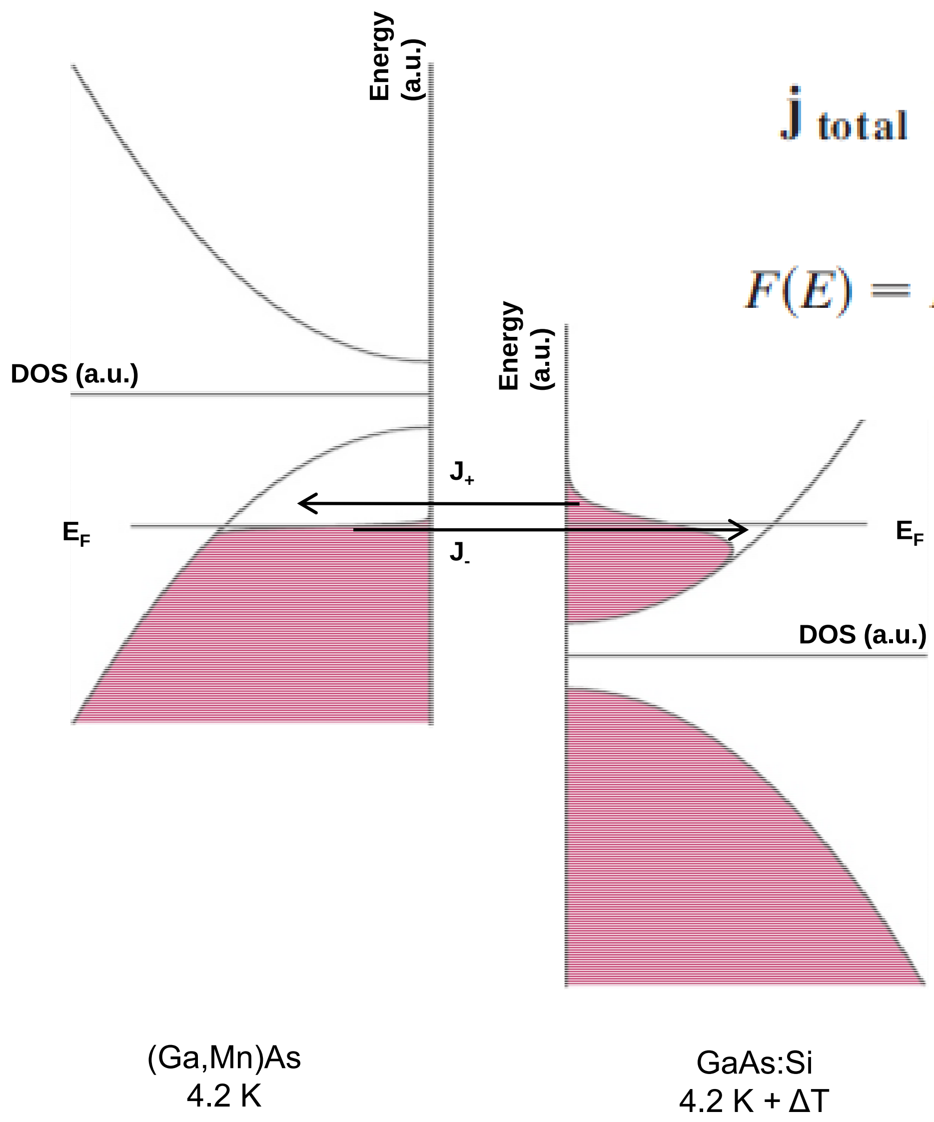


T. Naydenova et al., Phys. Rev. Lett. 107, 197201 (2011)

Model (originally for Fe/GaAs):

R. Cowburn, S. Gray, J. Ferré, J. Bland, and J. Miltat. *J. Appl. Phys.*, Vol. 78, p. 7210, 1995.

Thermopower Model and Simulation



$$j_{total} = A \int_{E_{C,GaAs:Si}}^{E_{V,(Ga,Mn)As} - eV_{th}} F(E)dE$$

$$F(E) = D_{GaAs:Si}(E) \cdot D_{(Ga,Mn)As}(E - eV_{th}) \times [f_{GaAs:Si}(E) - f_{(Ga,Mn)As}(E - eV_{th})]$$

Signal amplitude fully
in agreement with band model

$$S \simeq 0.4\ \mu V/K$$

Conclusions

- Current heating is a flexible technique for thermoelectric measurements on nanostructures. Avoids phonon drag, substrate effects.
- Many detailed investigations of quantum dot transport
- First observation of Kondo thermopower on a single impurity
- Discovered TAMT in a n-GaAs/(Ga,Mn)As junction

Collaborators:
Stefan Möller, Sandra Godijn, Ralph Scheibner, Tsvetelina Naydenova, Holger Thierschmann.

Hartmut Buhmann, Charles Gould

Funding: DFG