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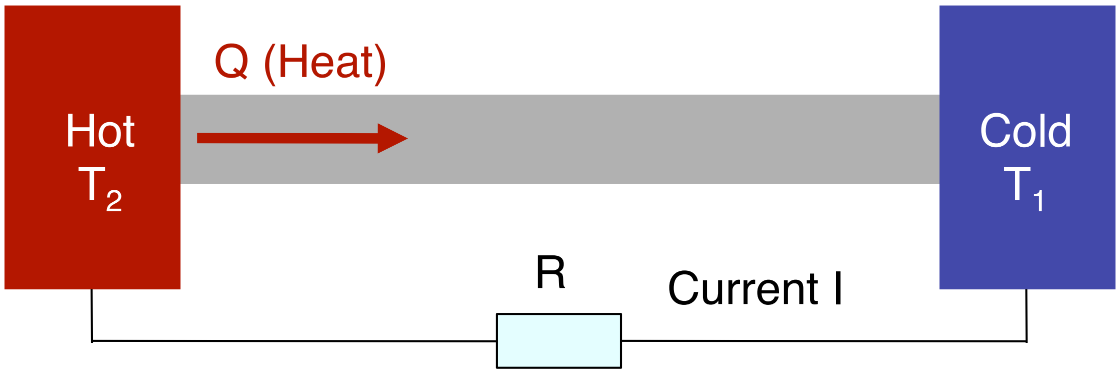
Realization of an efficient quantum-dot heat engine

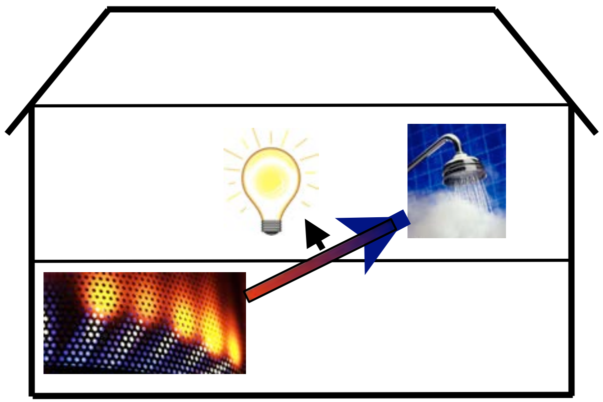
HEINER LINKE

NanoLund and Solid State Physics, Lund University, Sweden

Energy Conversion in the Quantum Regime, 27 August 2019, ICTP,Trieste

Thermoelectrics





- Low parasitic heat conduction by electrons (κ_{el}) and phonons (κ_{ph}).
- High Seebeck coefficient $S = \Delta V/\Delta T$
- Little Joule heating (high conductivity σ)

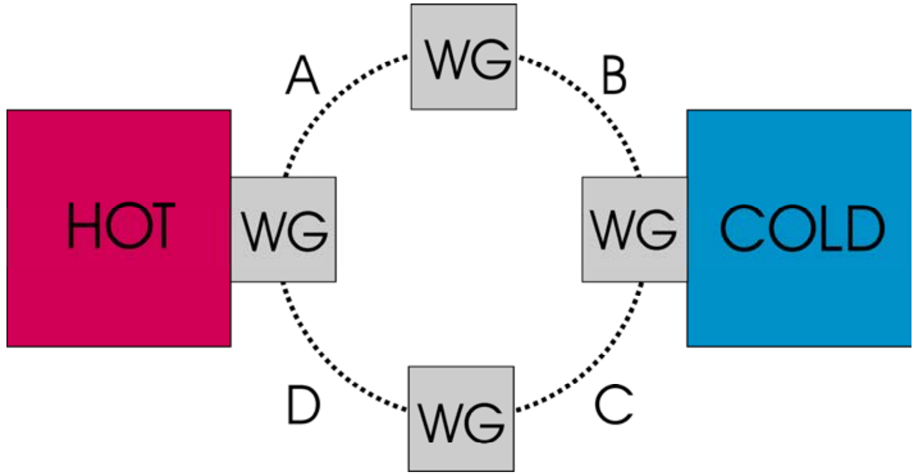
Figure of merit:

$$Z = \frac{S^2\sigma}{\kappa_e + \kappa_{ph}}$$

Power factor

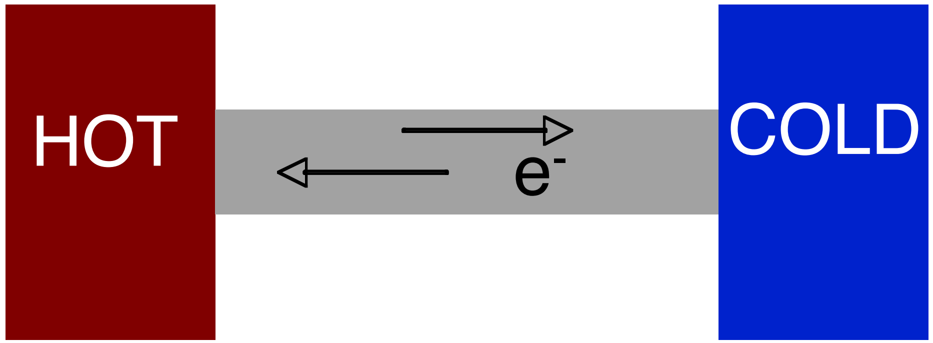
Fundamental efficiency limit of thermoelectrics?

Classic, cyclic Carnot engine:
Working gas (WG) in contact with only one heat reservoir at a time.



$$\eta_C = 1 - \frac{T_C}{T_H}$$

Thermoelectric:
In contact with both reservoirs at all times.



$$Z = \frac{S^2\sigma}{\kappa_e + \kappa_{ph}}$$

Outline

- Energy-filtering and energy-specific equilibrium in thermoelectrics
- Realizing “the best thermoelectric”: quantum-dot heat engines
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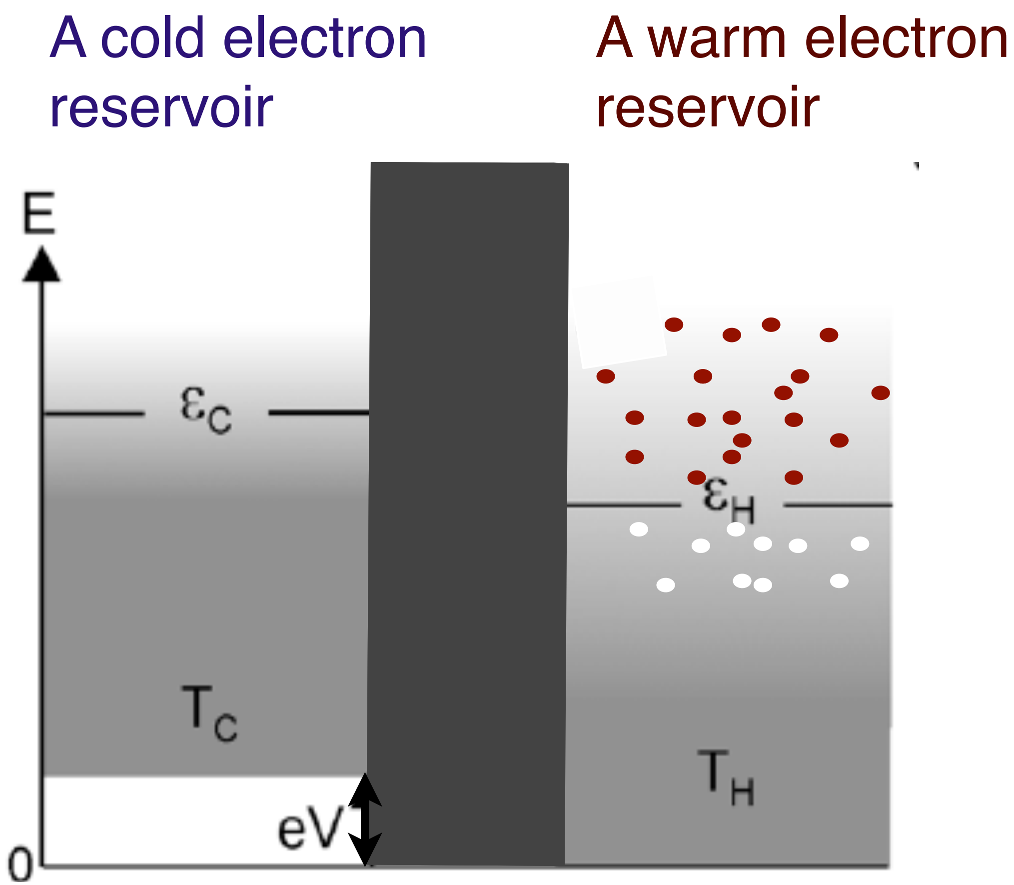
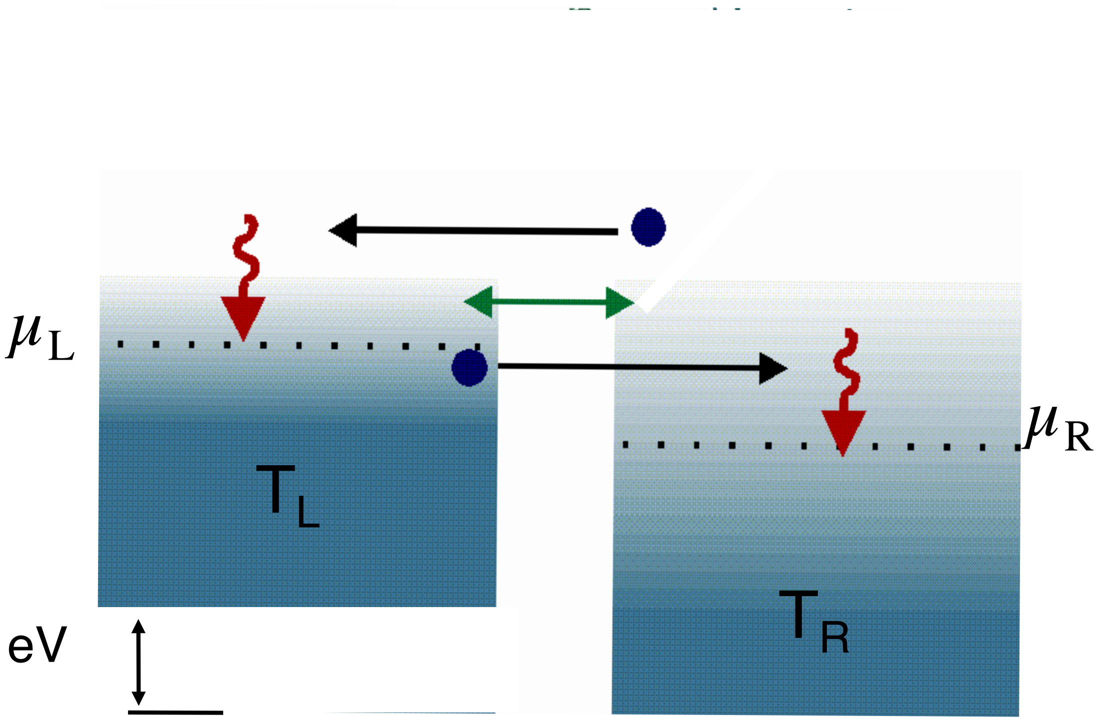
- Single-molecule “quantum dots”
- Application of energy filtering to hot-carrier solar cells

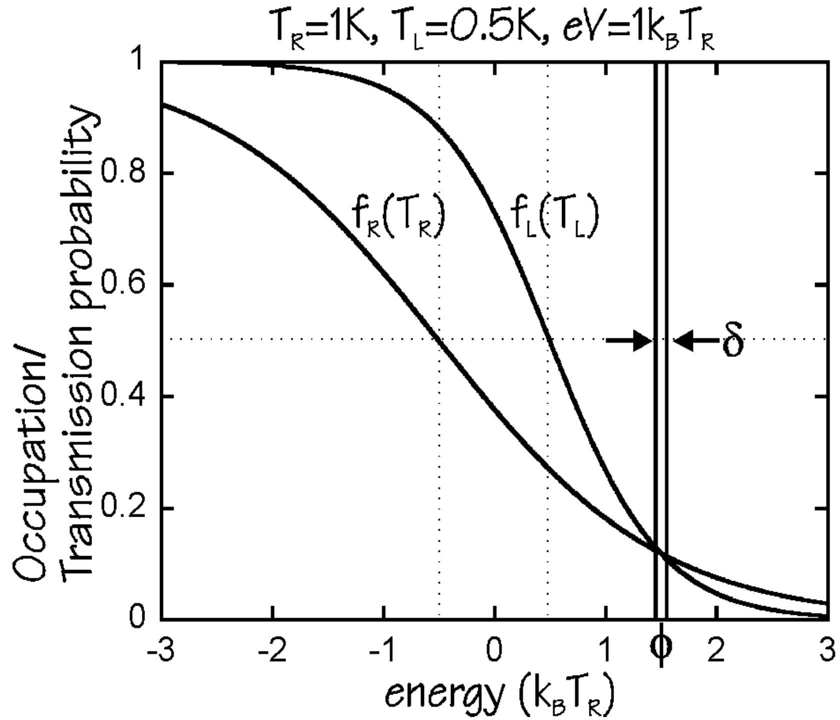
Fundamental elements of thermoelectrics



A bias voltage
to do work *against.*

Reversible electron transfer





Transfer of one electron
at energy ε from L to R:

$$\Delta S = \frac{-(\varepsilon - \mu_L)}{T_L} + \frac{(\varepsilon - \mu_R)}{T_R}$$

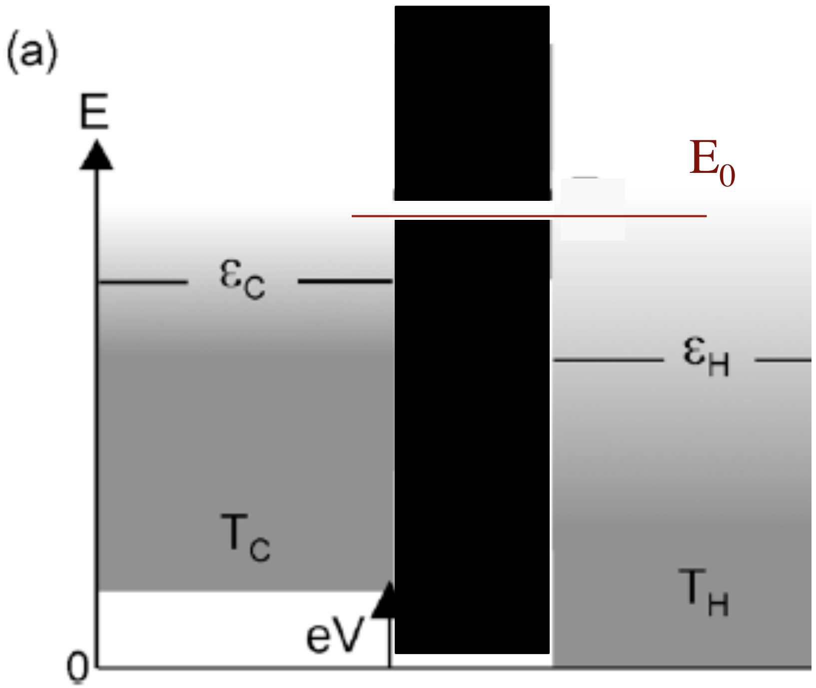
$$\Delta S = 0 \quad for \quad \varepsilon = \left(\frac{\mu_L T_R + \mu_R T_L}{T_R - T_L} \right)$$

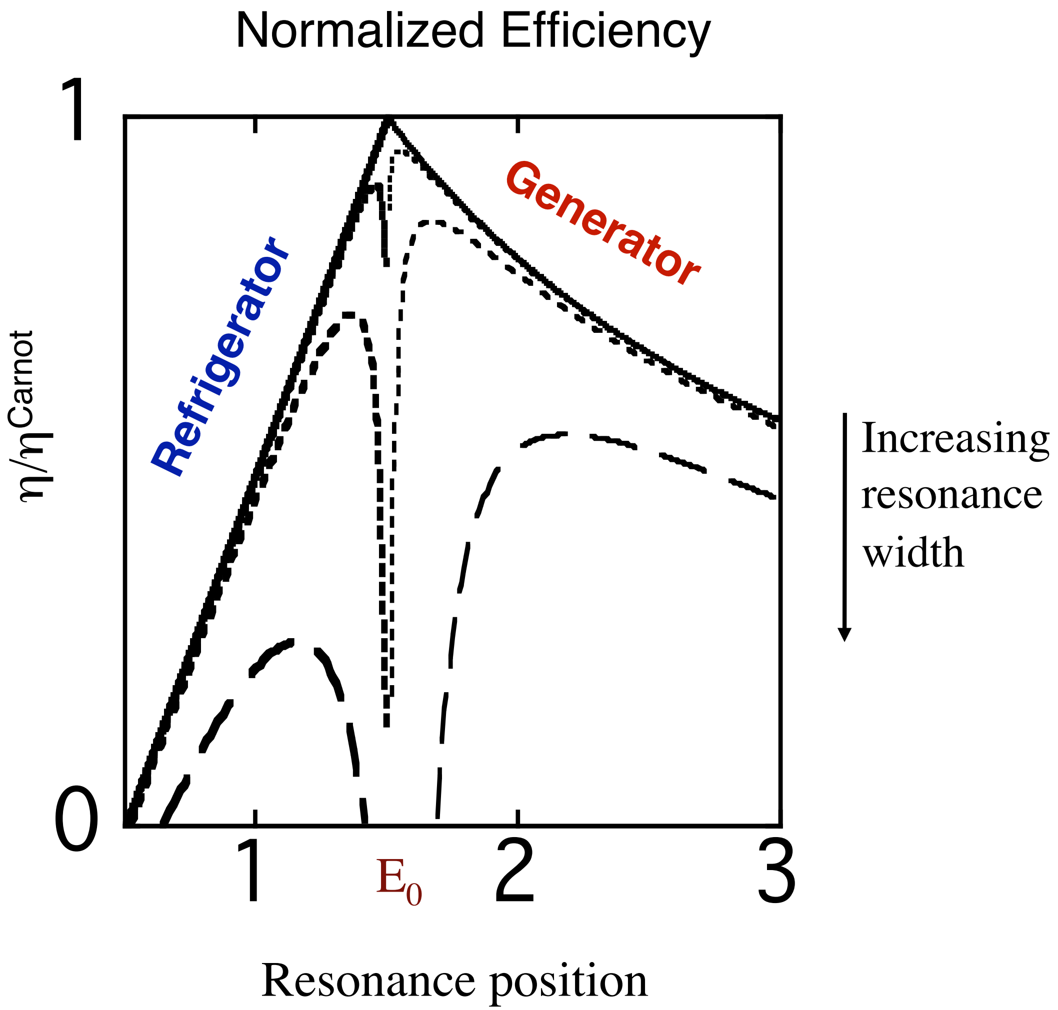
“Energy-specific equilibrium”

T. E. Humphrey and H. Linke, PRL **89,** 116801 (2002)

T. E. Humphrey, H Linke, PRL **94**, 096601 (2005)

Power generation or Refrigeration
with tuneable efficiency and power





T. E. Humphrey and H. Linke, PRL **89,** 116801 (2002)

The "best thermoelectric"

Proc. Natl. Acad. Sci. USA
Vol. 93, pp. 7436–7439, July 1996
Applied Physical Sciences

The best thermoelectric

G. D. MAHAN*† AND J. O. SOFO‡

*Department of Physics and Astronomy, The University of Tennessee, Knoxville, TN 37996-1200; †Solid State Division, Oak Ridge National Laboratory, P.O. Box 2008, Oak Ridge, TN 37831-6030; and ‡Instituto Balseiro, Centro Atomico Bariloche, (8400) Bariloche, Argentina

Contributed by G. D. Mahan, May 20, 1996

ABSTRACT What electronic structure provides the largest figure of merit for thermoelectric materials? To answer that question, we write the electrical conductivity, thermopower, and thermal conductivity as integrals of a single function, the transport distribution. Then we derive the mathematical function for the transport distribution, which gives the largest figure of merit. A delta-shaped transport distribution is found to maximize the thermoelectric properties. This result indicates that a narrow distribution of the energy of the electrons participating in the transport process is needed for maximum thermoelectric efficiency. Some possible realizations of this idea are discussed.

$$Z = \frac{S^2\sigma}{\kappa_e + \kappa_{ph}}$$

Figure of merit

Efficiency at maximum power: Curzon-Ahlborn efficiency

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$$\eta_c = 1 - T_c / T_h$$

Carnot efficiency requires reversible operation, which is equivalent to zero power output.

$$\eta_{CA} = 1 - \sqrt{T_c / T_h}$$

Curzon-Ahlborn efficiency describes the efficiency of an ideal Carnot engine operated at maximum power (neglecting dissipation in reservoirs)

$$\eta_{CA} = 1 - \sqrt{T_c / T_h} = \eta_c / 2 + \eta_c^2 / 8 + \cdots$$

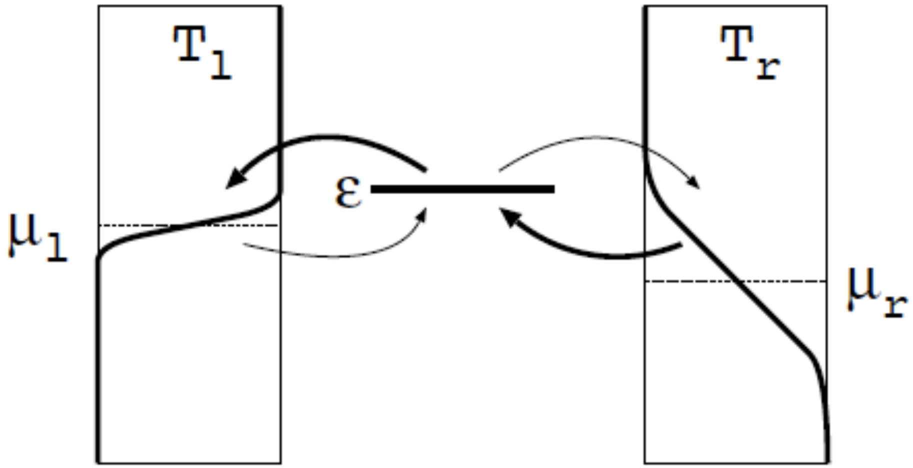
II Novikov, J Nuclear Energy **7**, 125 (1954).

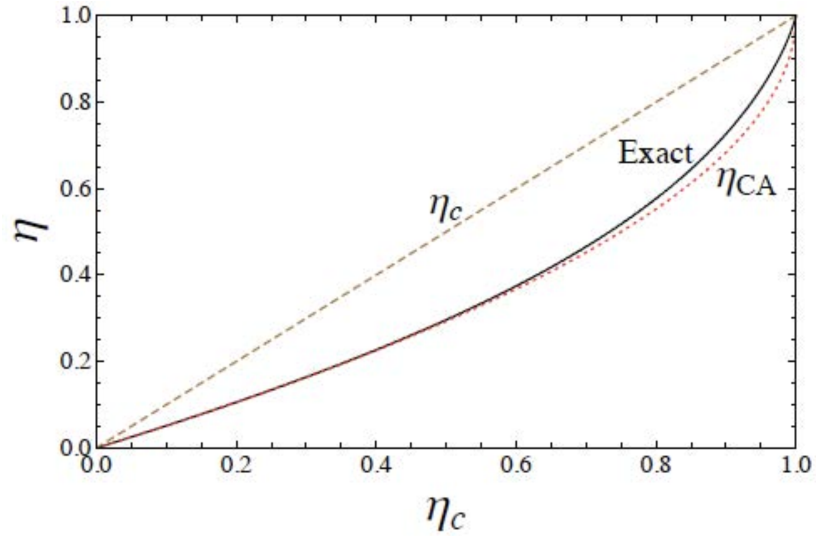
F. Curzon and B. Ahlborn, Am. J. Phys. **43**, 22 1975.

Thermoelectric efficiency at maximum power in a quantum dot

Massimiliano Esposito* and Katja Lindenberg
Department of Chemistry and Biochemistry and Institute for Nonlinear Science, University of California, San Diego, La Jolla, CA 92093-0340, USA

Christian Van den Broeck
Hasselt University, B-3590 Diepenbeek, Belgium





$$\eta = \frac{\eta_c}{2} + \frac{\eta_c^2}{8} + \frac{[7 + \mathrm{csch}^2(a_0/2)]}{96}\eta_c^3 + \mathcal{O}(\eta_c^4)$$

Esposito, Lindenberg, van den Broeck, PRL **102**, 130602 (2009)

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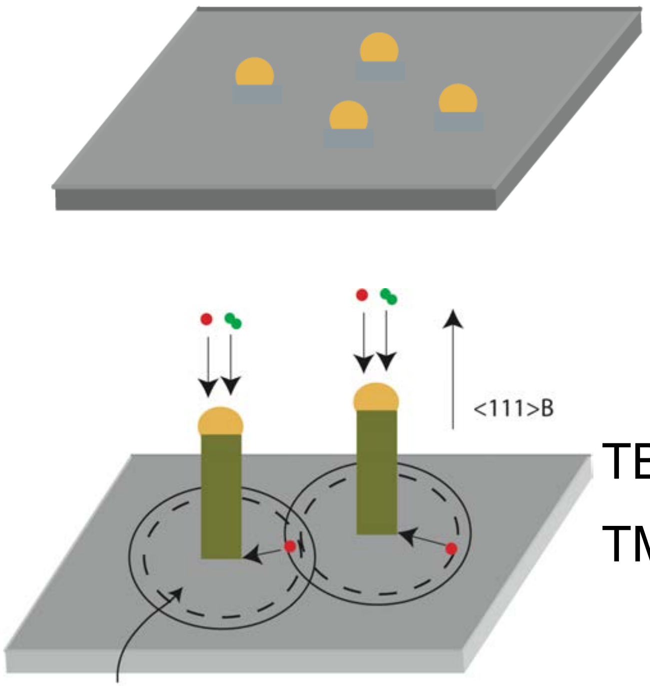
Epitaxially grown nanowires, e.g. InAs/InP

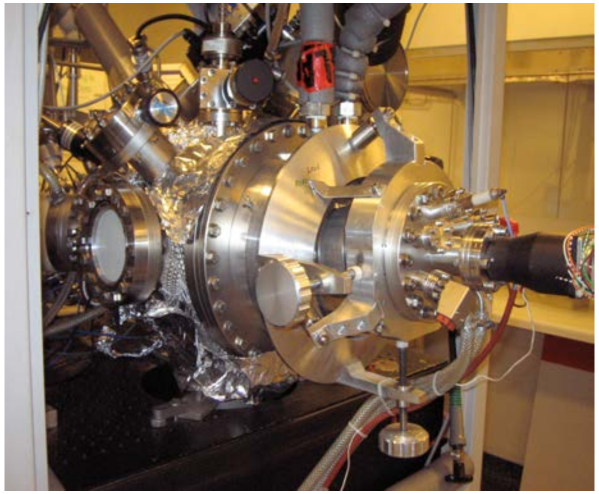


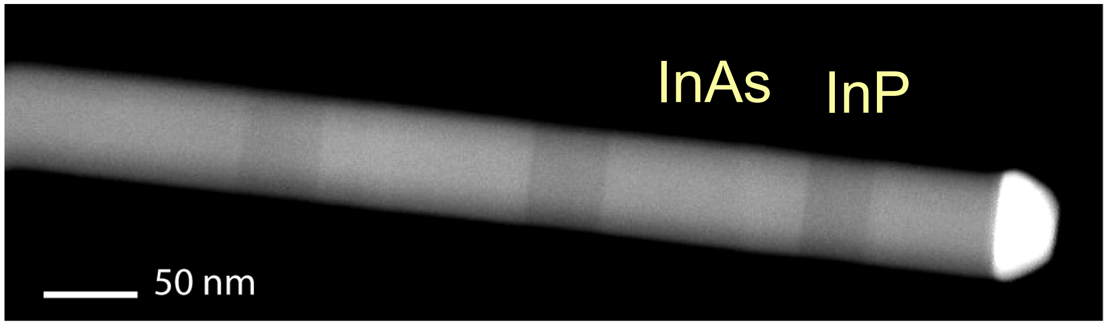
TBAs (group-V)

TMIn (group-III)

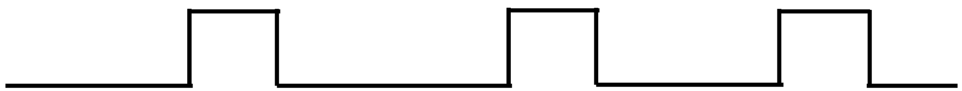
Substrate surface
InAs (111)B

CBE





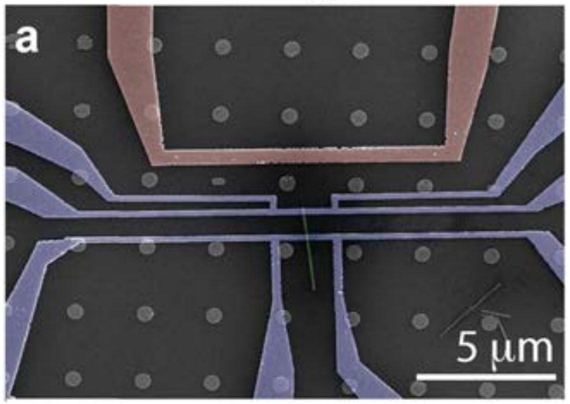
Conduction band edge

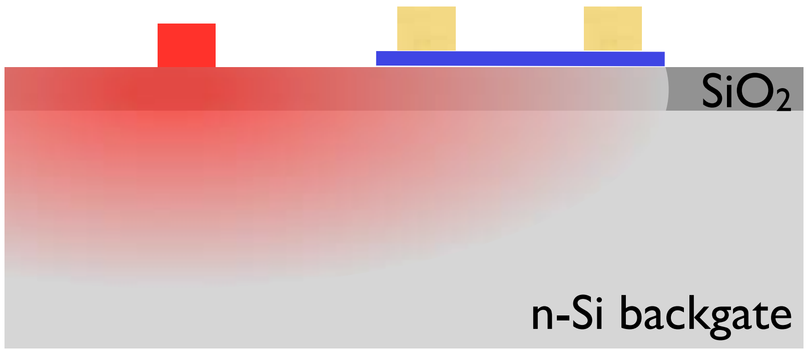


Ann Persson, Linus Fröberg

Applying a temperature gradient along a nanowire

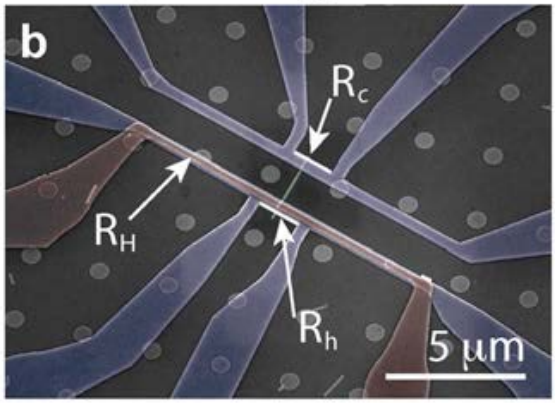
Traditional side-heater

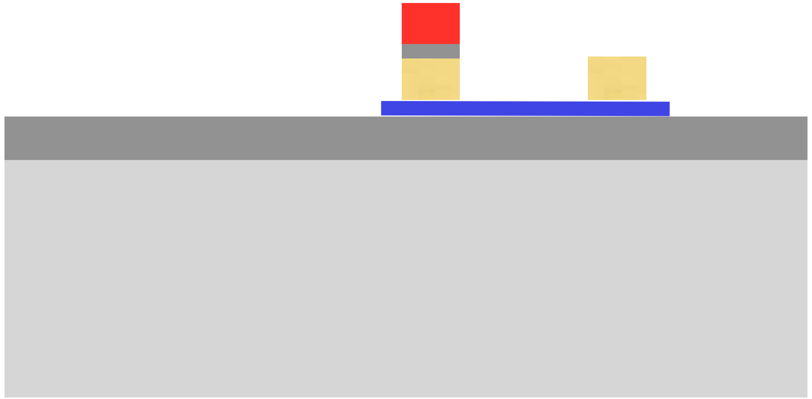




Substantial global device heating limits use for low-temperature experiments

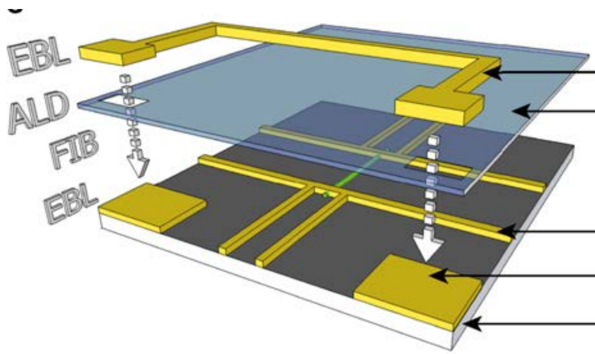
Local top-heater

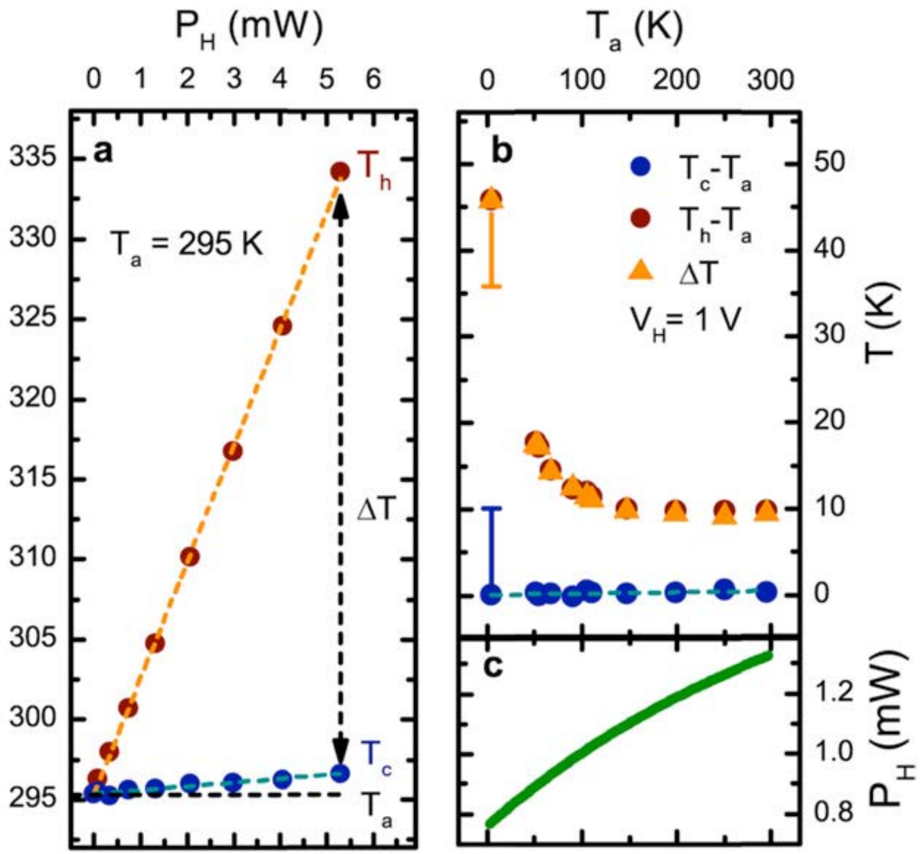




Local, direct heating of the warm contact without electrical interference.

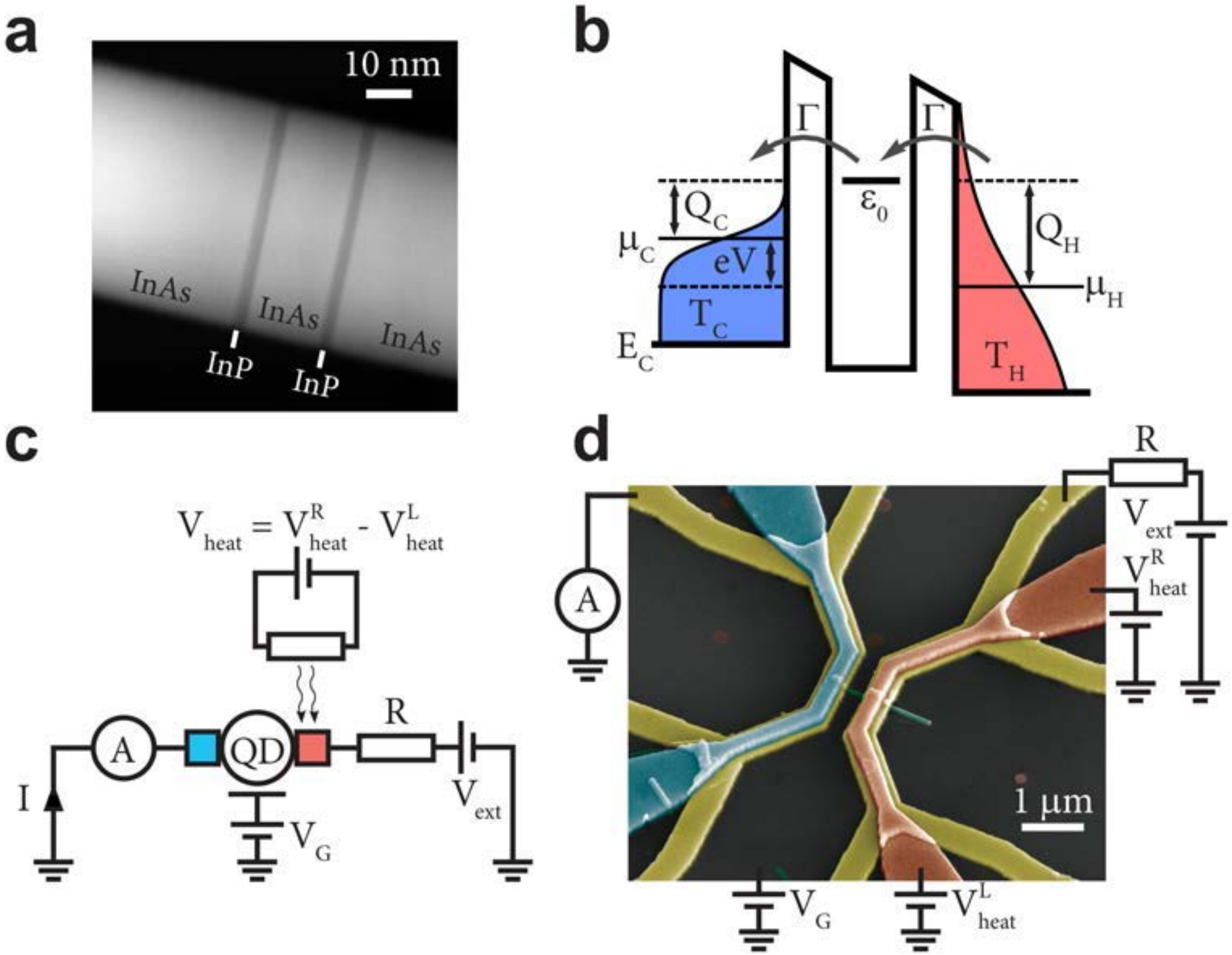
Top-heaters to enable high ΔT with minimal heating





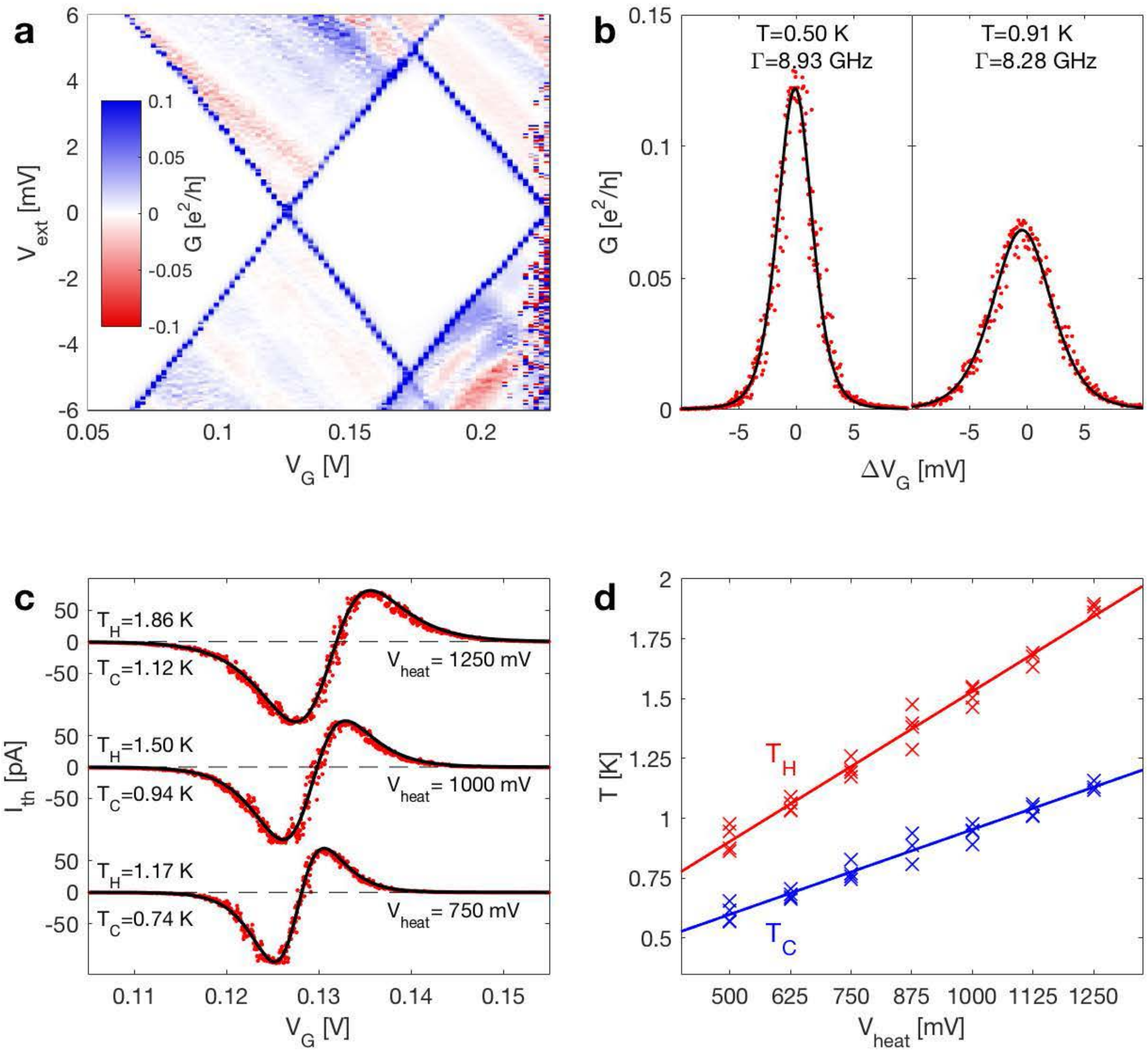
J. Gluschke et al
Nanotechnology **25**, 385704 (2014)

Quantum-dot heat engine: device



Artis Svilans

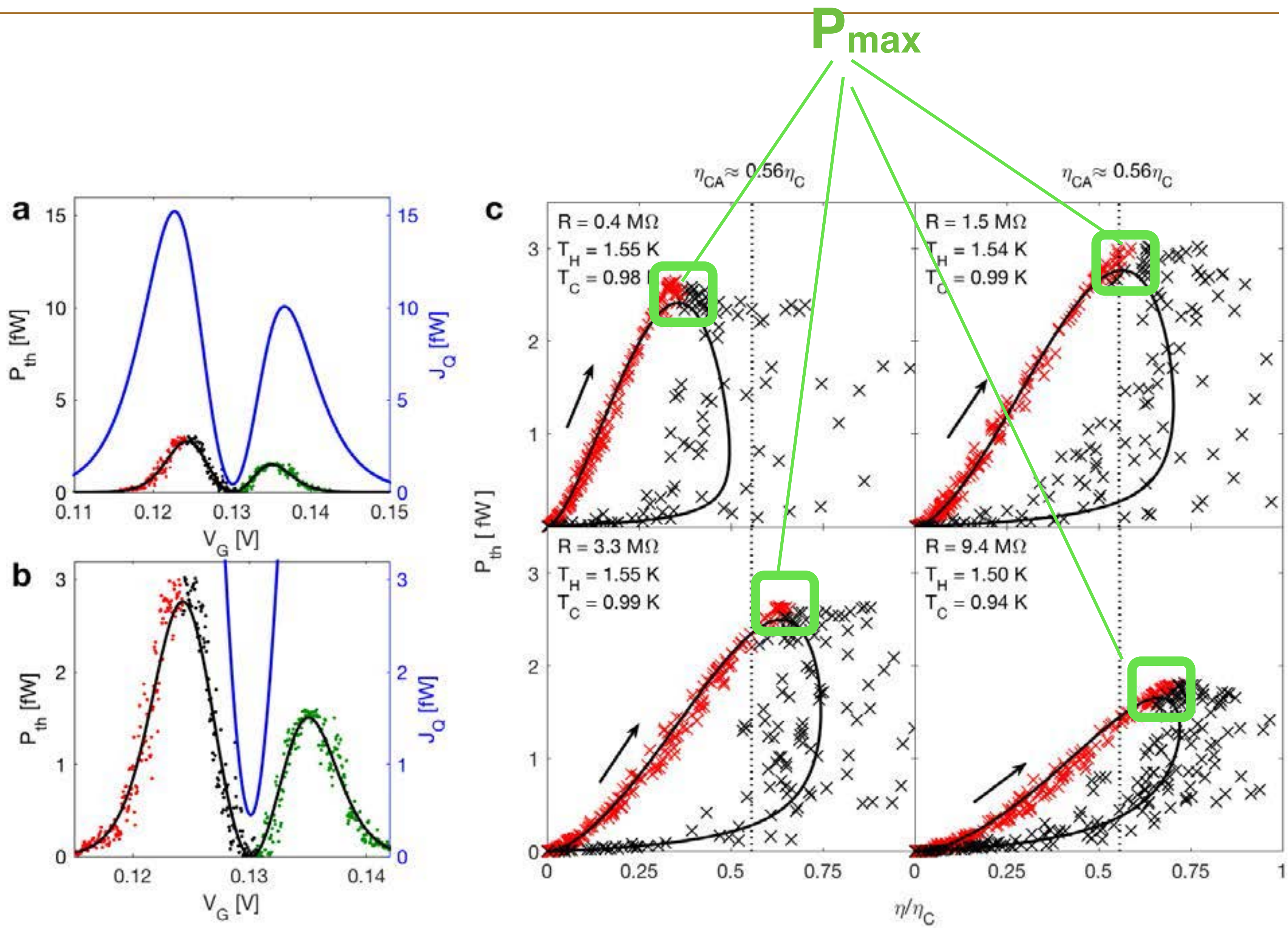
Quantum-dot heat engine: characterisation



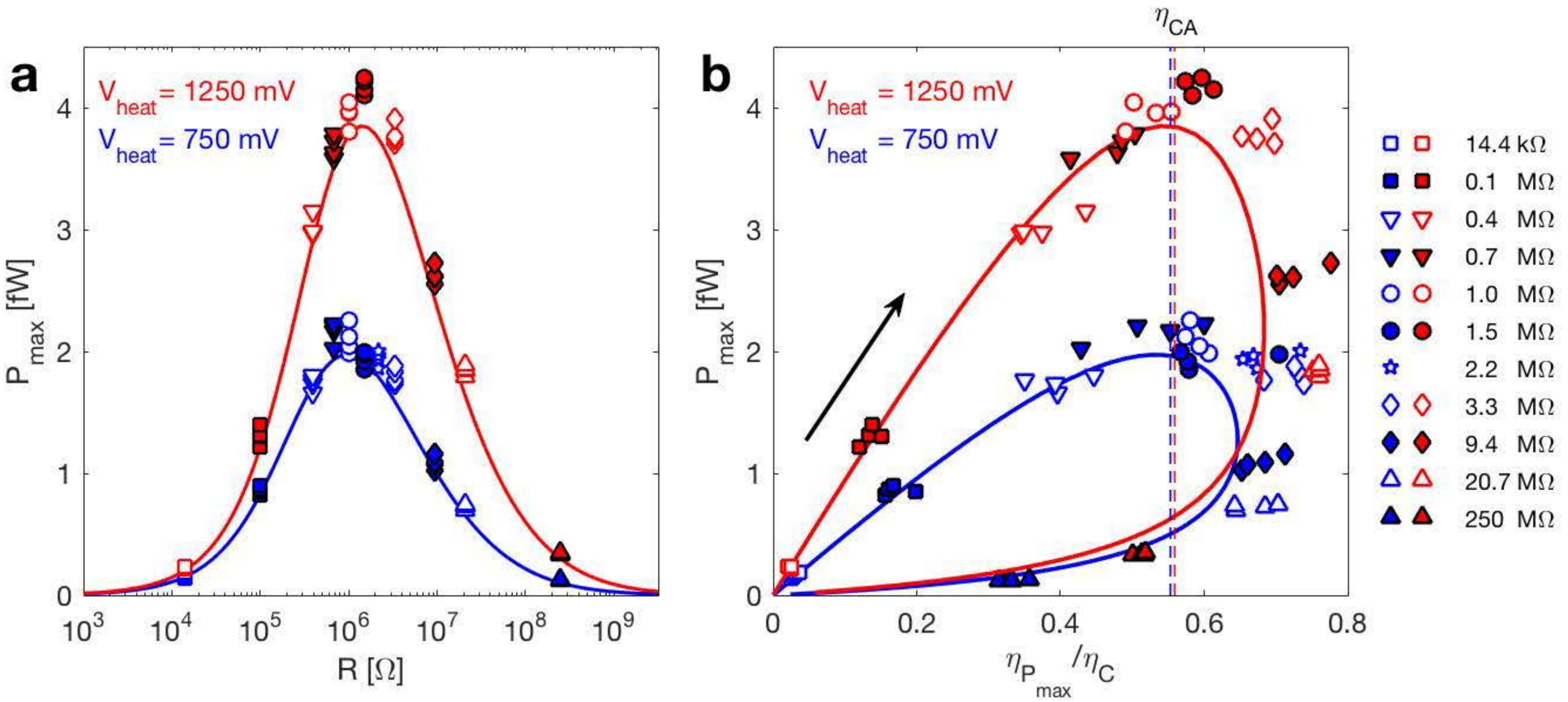
Artis Svilans

Martin Josefsson

Quantum-dot heat engine: performance



Quantum-dot heat engine: 70% of Carnot efficiency demonstrated



Quantum-dot heat engine achieves Curzon-Ahlborn efficiency at maximum power and about 70 % of Carnot efficiency with finite power output

M. Josefsson, A. Svilans, et al. Nature Nanotechnology **13**, 920 (2018)

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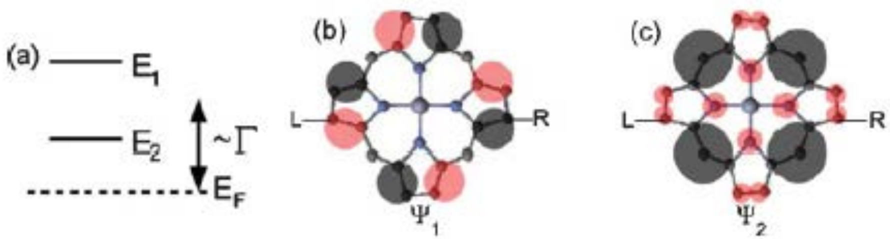
- Application of energy filtering to hot-carrier solar cells

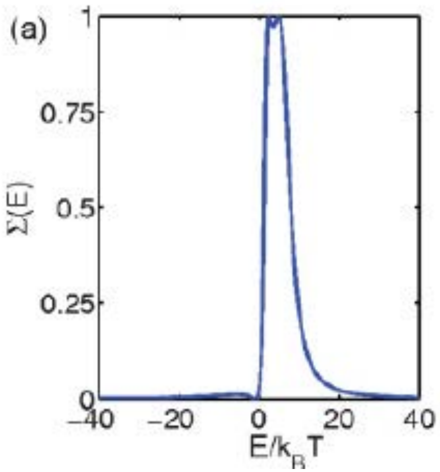
Higher maximum power by using interference

PHYSICAL REVIEW B 84, 113415 (2011)

Increasing thermoelectric performance using coherent transport

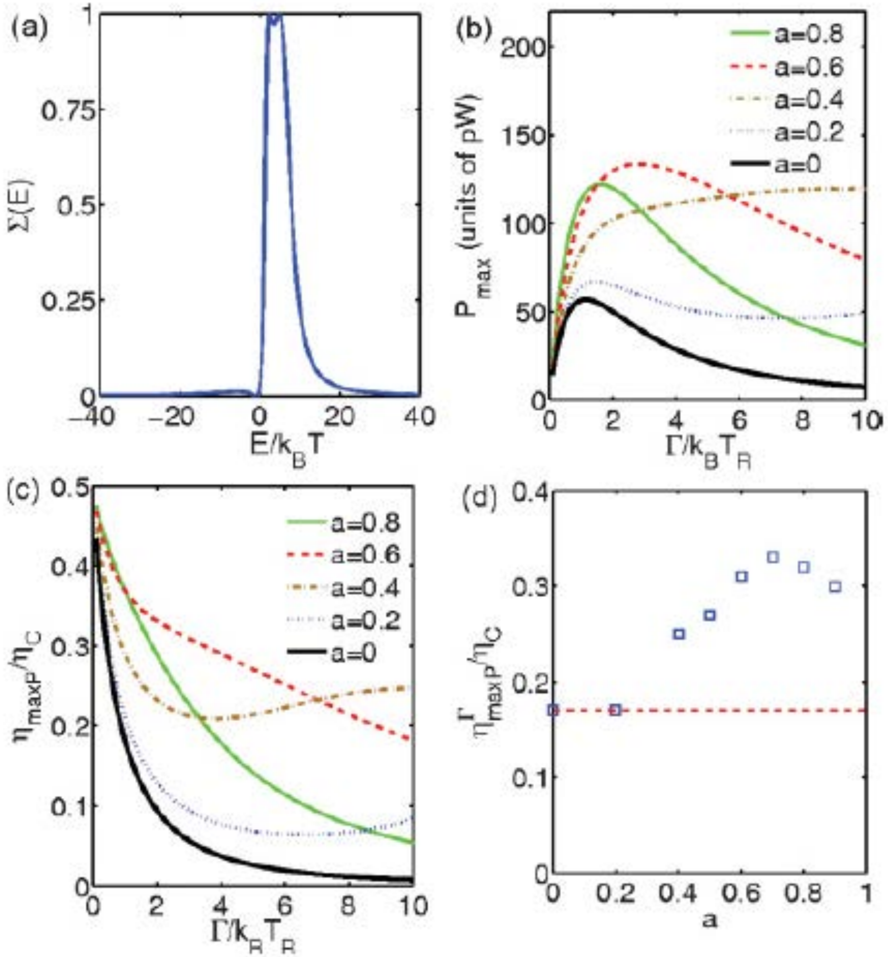
O. Karlström,[1,*] H. Linke,[1] G. Karlström,[2] and A. Wacker[1]

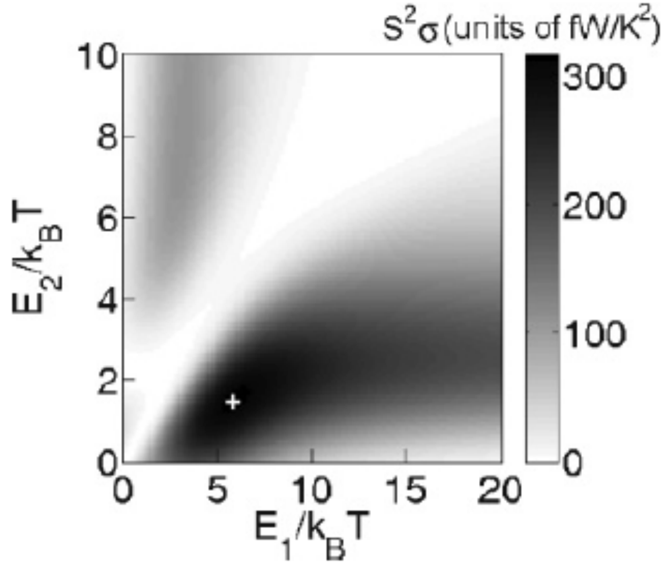




$$\Sigma(E) = \Gamma^2 \left| \frac{1}{E - E_1 + i\Gamma} - \frac{a^2}{E - E_2 + ia^2\Gamma} \right|^2$$

Higher maximum power by using interference





$$\Sigma(E) = \Gamma^2 \left| \frac{1}{E - E_1 + i\Gamma} - \frac{a^2}{E - E_2 + ia^2\Gamma} \right|^2$$

First step: interference effects yield measurable difference in thermopower

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NANO LETTERS

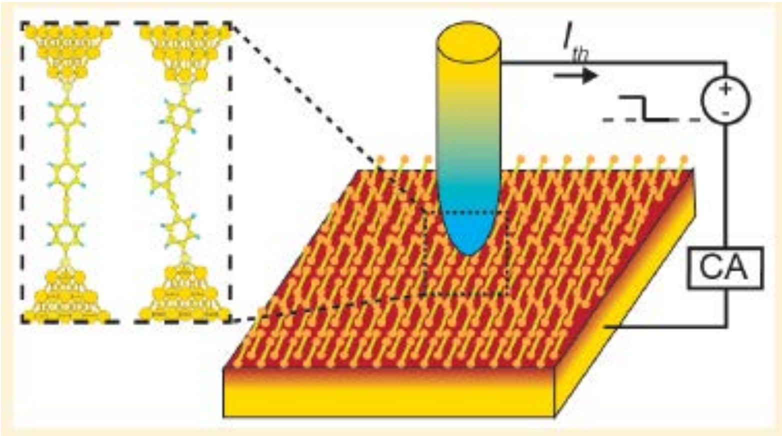
Letter

pubs.acs.org/NanoLett

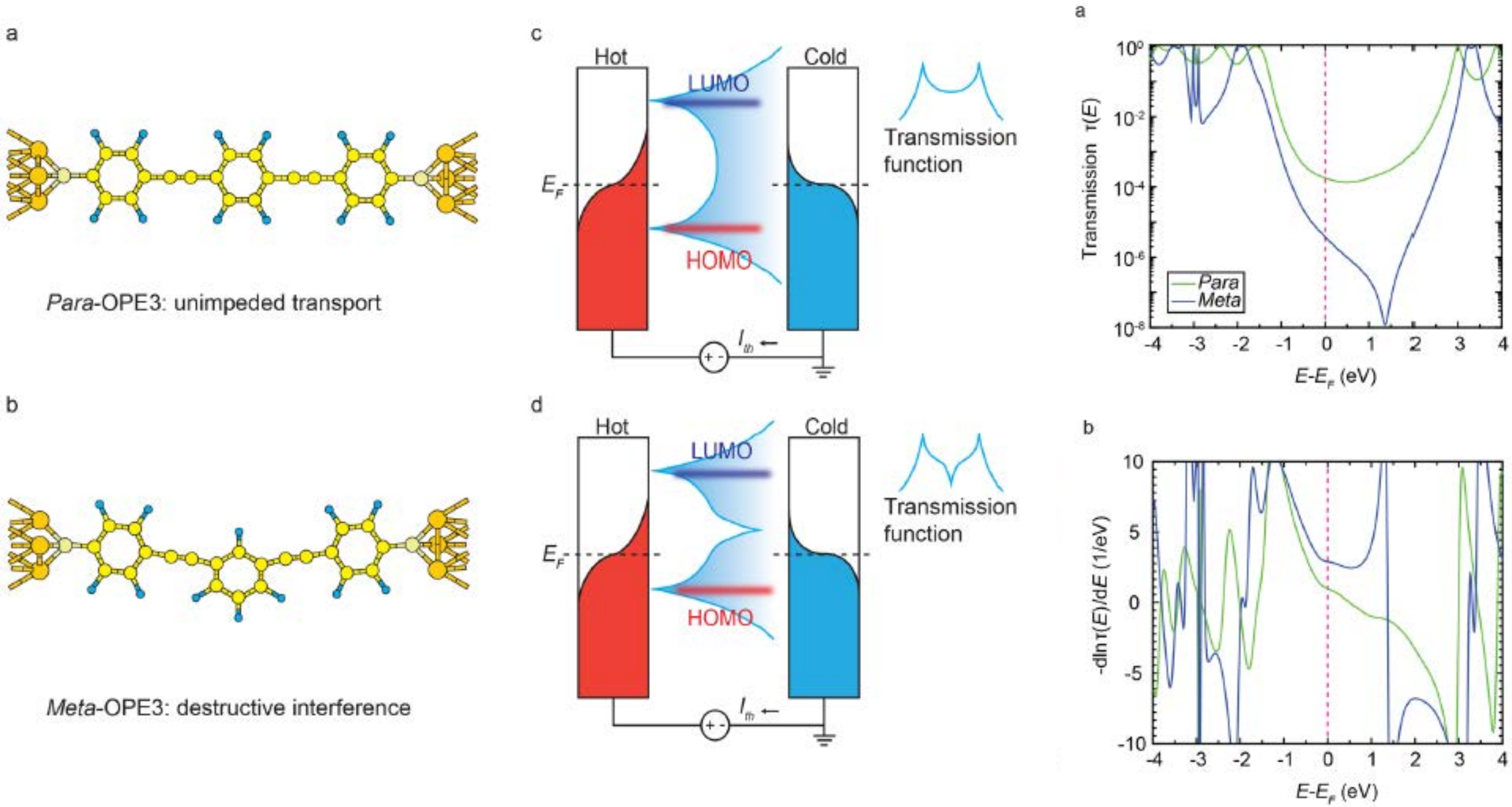
Influence of Quantum Interference on the Thermoelectric Properties of Molecular Junctions

Ruijiao Miao,[†] Hailiang Xu,[‡,§] Maxim Skripnik,[∥,⊥] Longji Cui,[†] Kun Wang,[†] Kim G. L. Pedersen,[#,▽] Martin Leijnse,[‡,○] Fabian Pauly,[∥,⊥] Kenneth Wärnmark,[‡,§] Edgar Meyhofer,[*,†] Pramod Reddy,[*,†,◆] and Heiner Linke[*,‡,○]

August 2018



First step: interference effects yield measurable difference in thermopower



Higher thermopower S predicted in presence of destructive interference

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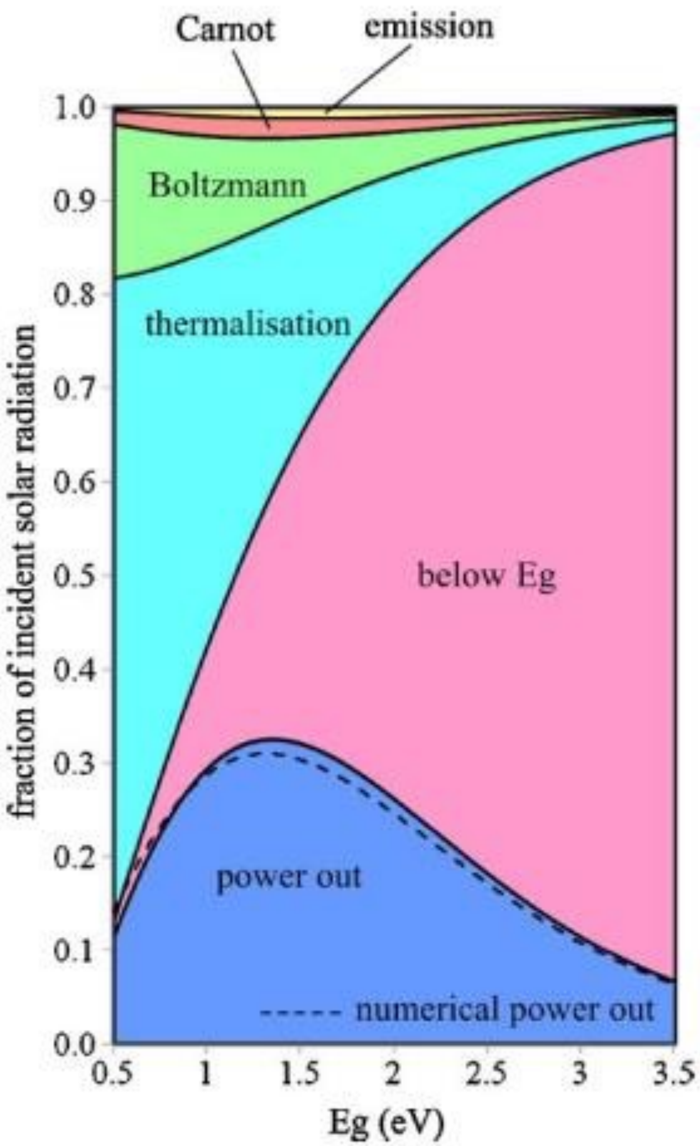
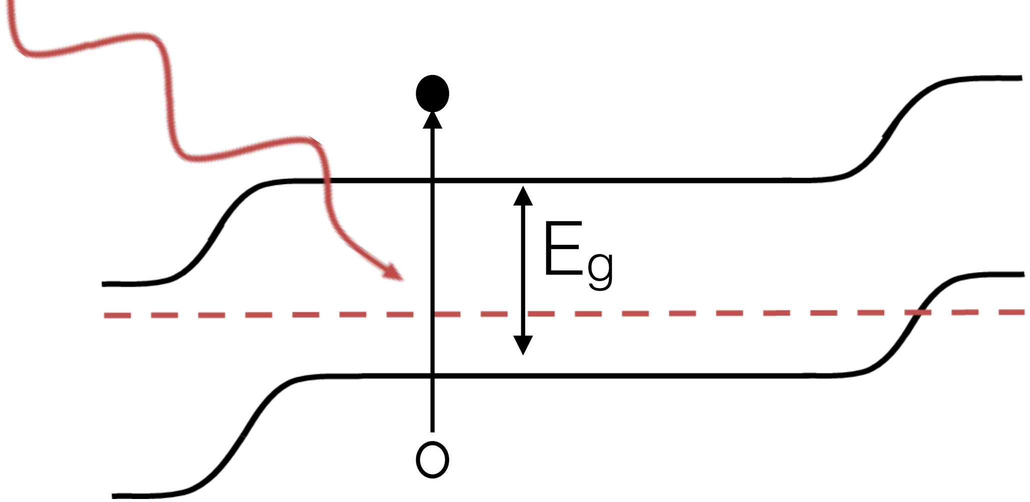


Figure 6. Intrinsic loss processes and hence, power out are shown to be dependent on E_g. All incident radiation is accounted for, illustrating why intrinsic loss mechanisms lead to fundamental limiting efficiency.



- Carrier cooling decreases the energy each carrier can provide to an external circuit.
- pn-junction solar cells of small bandgap materials are rarely made due to the magnitude of thermalisation losses.

L. C. Hirst and N. J. Ekins-Daukes, "Fundamental losses in solar cells," *Progr. in Photovolt.: Res. Appl.*, **19** 286–293 (2011)

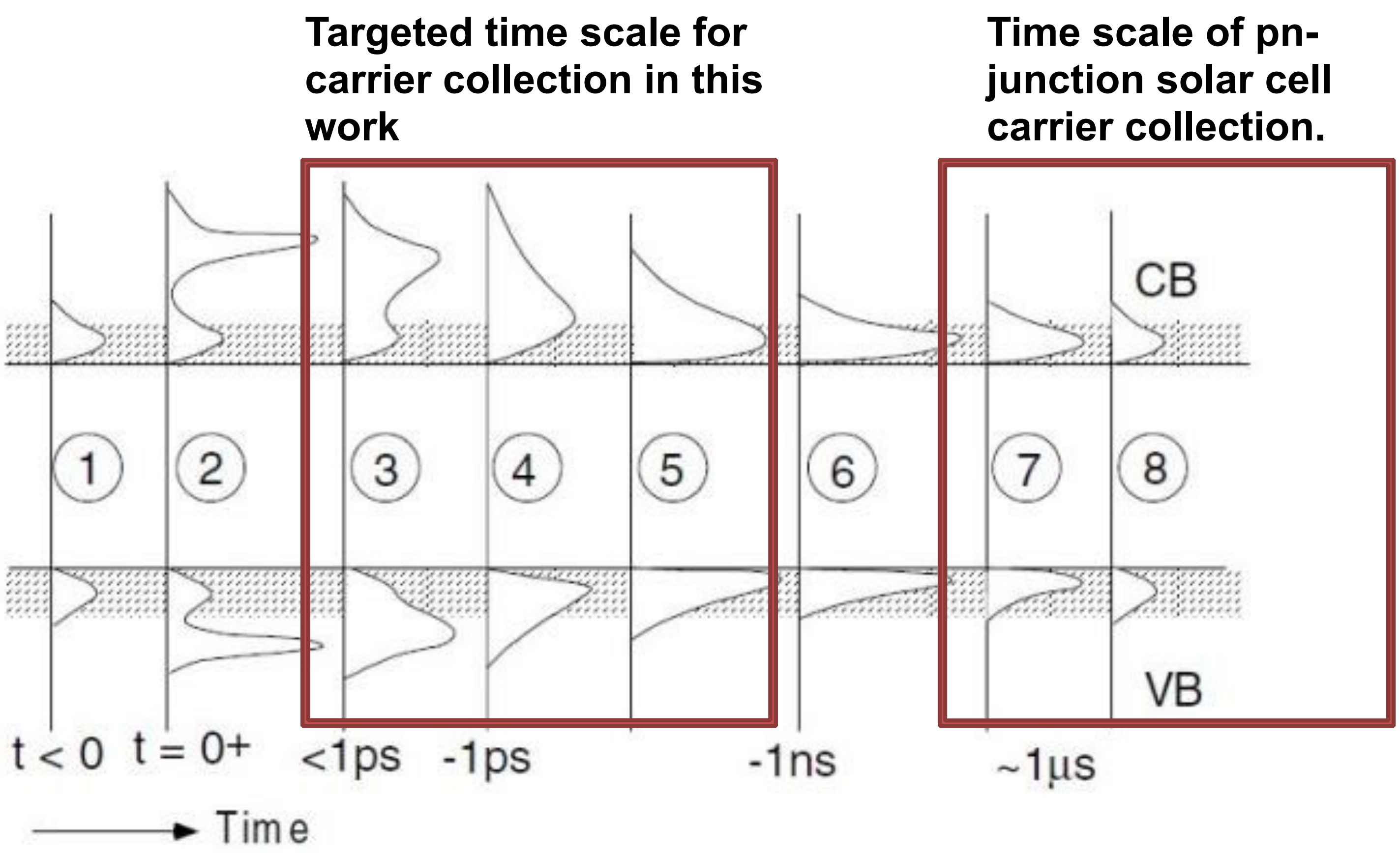
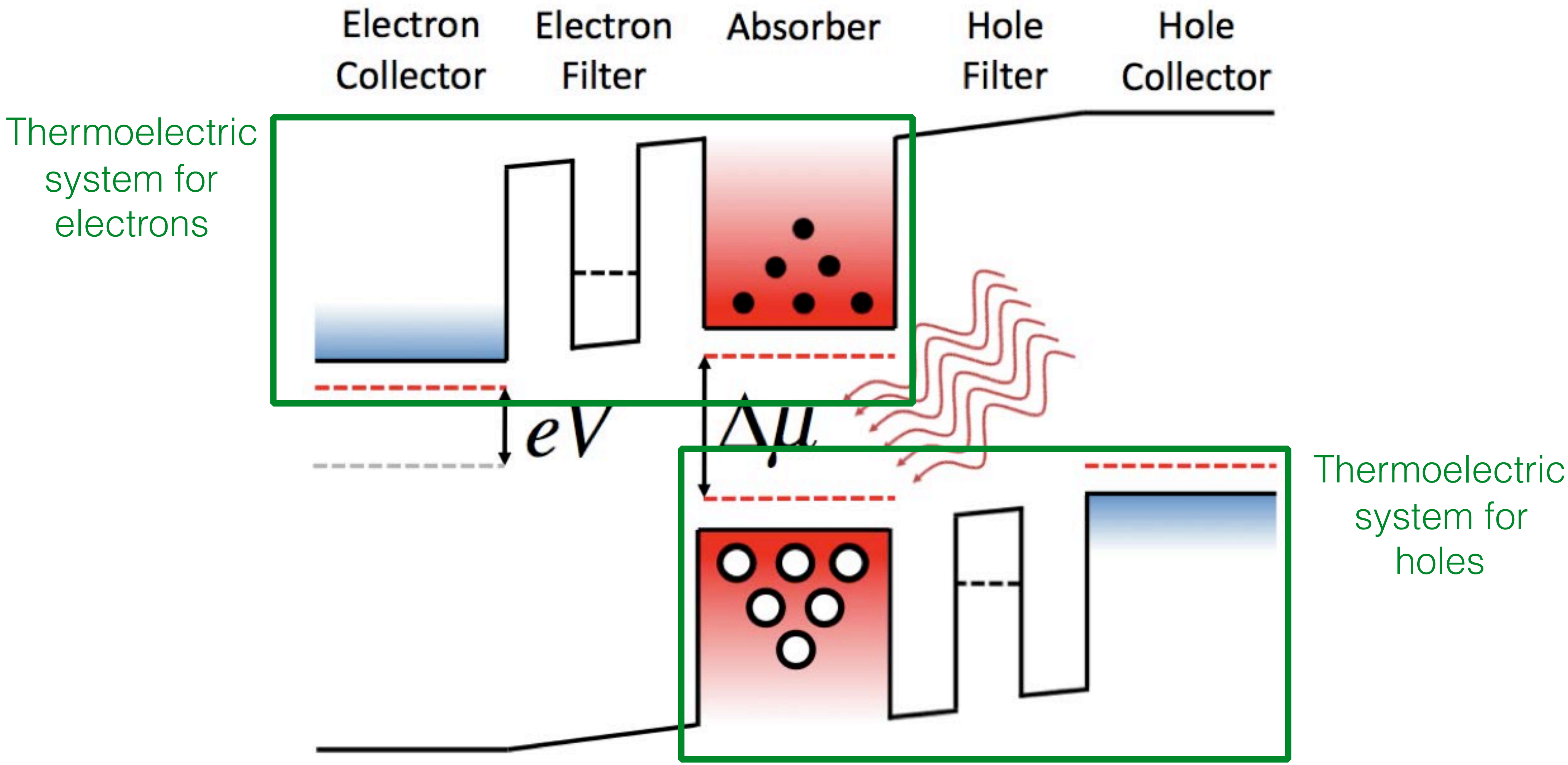


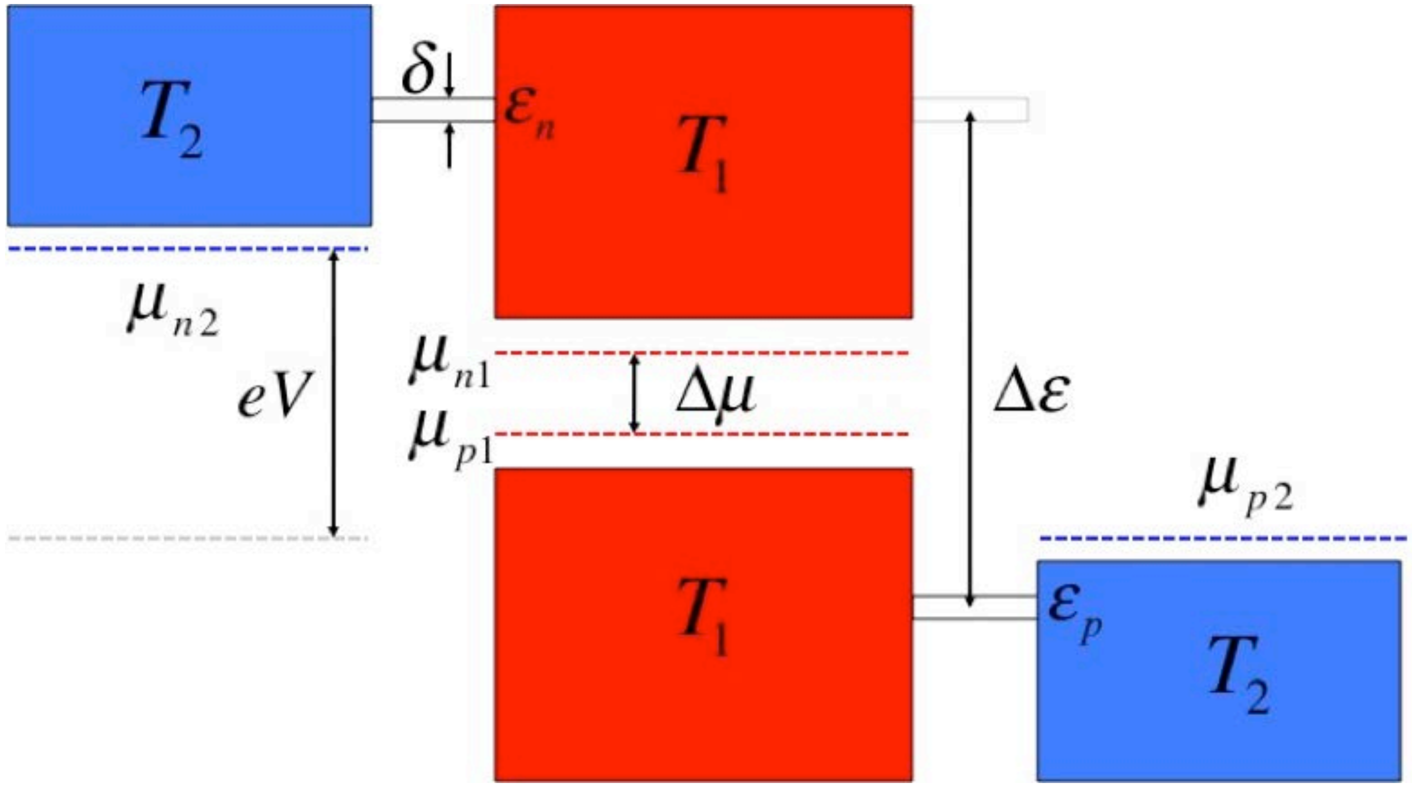
Fig. 6.2: Time evolution of electron and hole distributions in a semiconductor subject to a short, high intensity, monochromatic pulse of light from a laser: (1) Thermal equilibrium before pulse; (2) "coherent" stage straight after pulse; (3) carrier scattering; (4) thermalisation of "hot carriers"; (5) carrier cooling; (6) lattice thermalised carriers; (7) recombination of carriers; (8) return to thermal equilibrium.

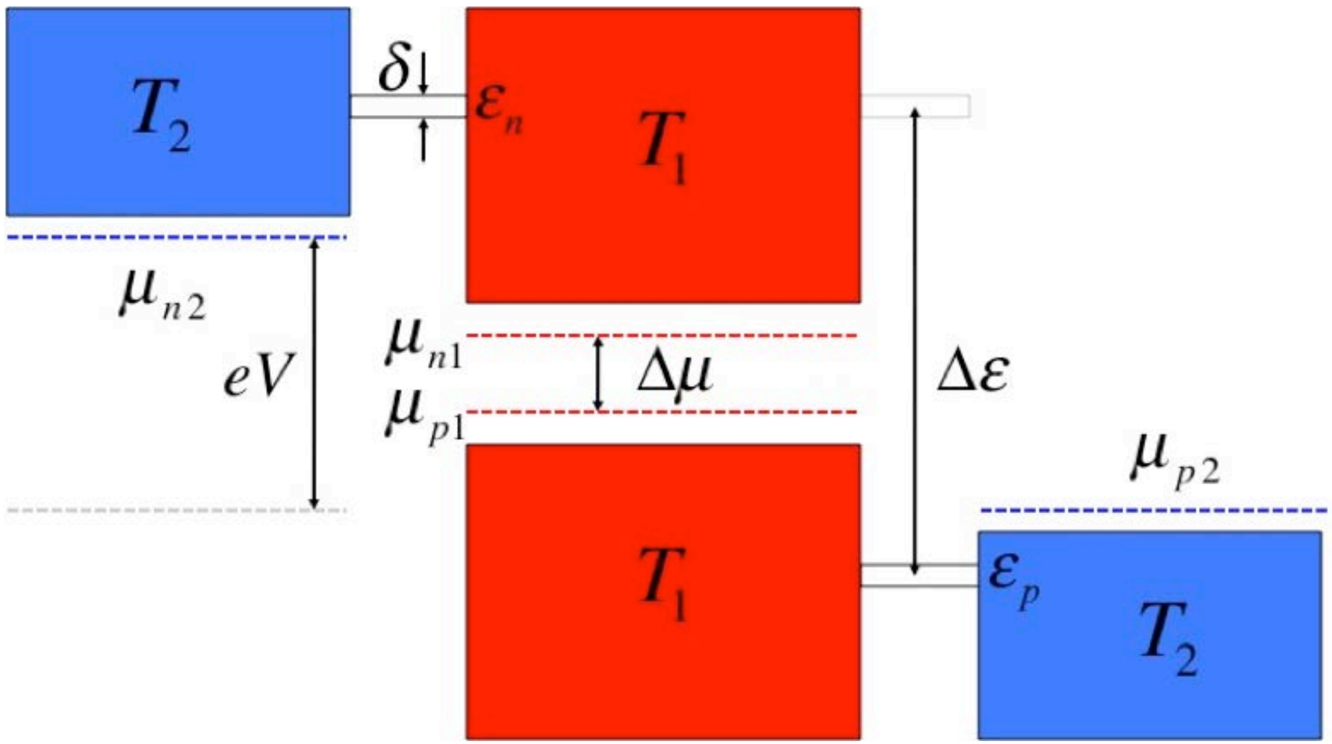
M. A. Green, Third Generation Photovoltaics. Heidelberg: Springer, 2006.

Basic idea of a hot-carrier cell: photothermoelectrics



Can a hot-carrier photovoltaic system be run reversibly?





$$\Delta S = \Delta S_{n1,ext} + \Delta S_{n2,inj} + \Delta S_{p1,ext} + \Delta S_{p2,inj}$$
$$= \frac{\Delta\varepsilon - eV}{T_2} - \frac{\Delta\varepsilon - \Delta\mu}{T_1}.$$

ΔS = 0 when $\dfrac{\Delta\varepsilon - \Delta\mu}{T_1} = \dfrac{\Delta\varepsilon - eV}{T_2}$

(equivalent to energy-specific equilibrium across both junctions)

S. Limpert, S. Bremner, H. Linke,
New J. Phys. (2015)

Open-circuit voltage

Steven Limpert

$$\Delta S = \Delta S_{n1,ext} + \Delta S_{n2,inj} + \Delta S_{p1,ext} + \Delta S_{p2,inj}$$
$$= \frac{\Delta\varepsilon - eV}{T_2} - \frac{\Delta\varepsilon - \Delta\mu}{T_1}.$$

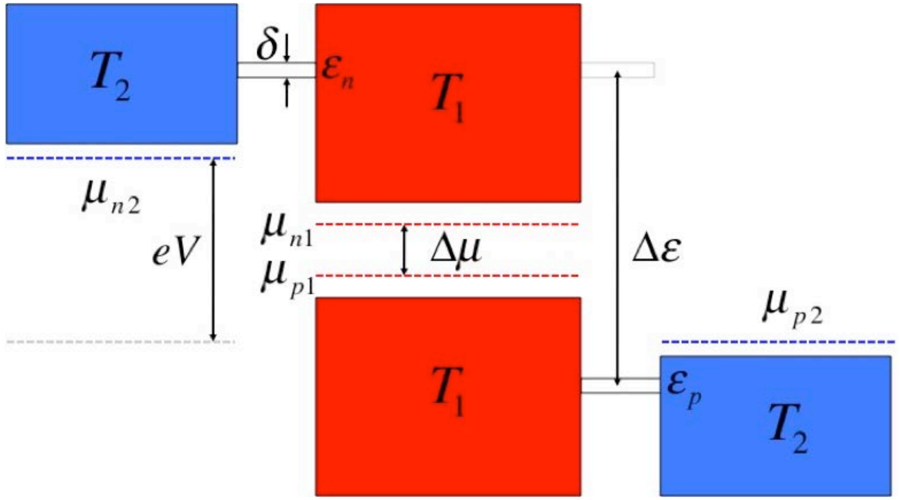
$$eV = \Delta\varepsilon\left(1 - \frac{T_2}{T_1}\right) + \Delta\mu\frac{T_2}{T_1} - T_2\Delta S.$$

$$= Q_1\eta_{Carnot} + \Delta\mu - T_2\Delta S.$$

Heat engine

Solar cell

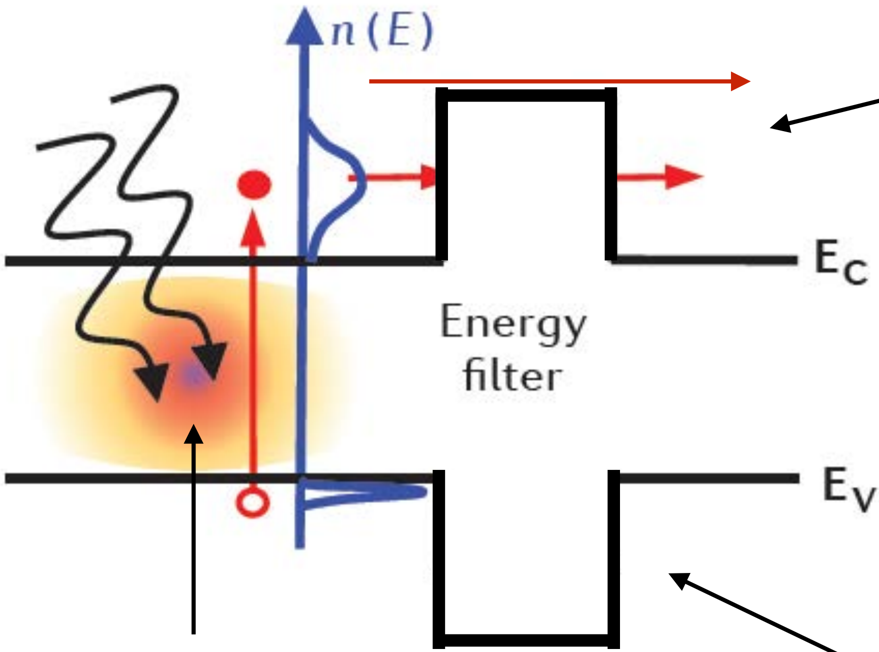
Explicit term describing the reduction of voltage due to irreversibility



$$Q_1 = Q_{n1} + Q_{p1} = (\varepsilon_n - \mu_{n1}) + (\mu_{p1} - \varepsilon_p) = \Delta\varepsilon - \Delta\mu$$

Basic idea for hot-carrier experiments

Heterostructure nanowire with small band gap and high electron-hole mass asymmetry (e.g. InAs/InP)

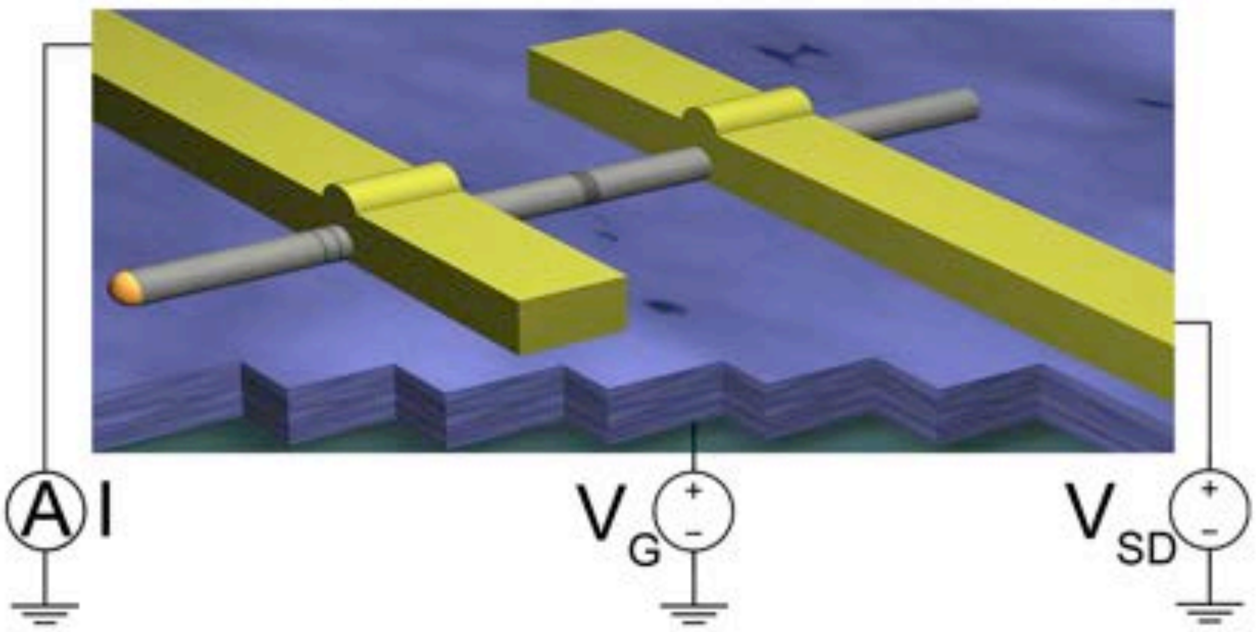


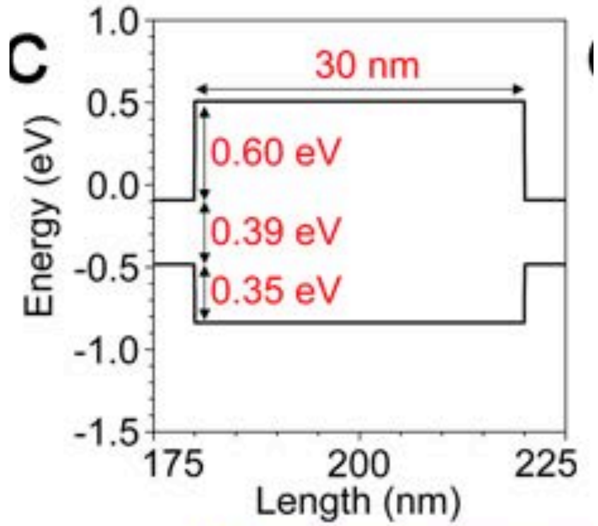
Energy filter (or thermionic barrier) for hot electrons

Local light absorption (photonic hot spot)

Block for holes (high mass -> small kinetic energy)

Device

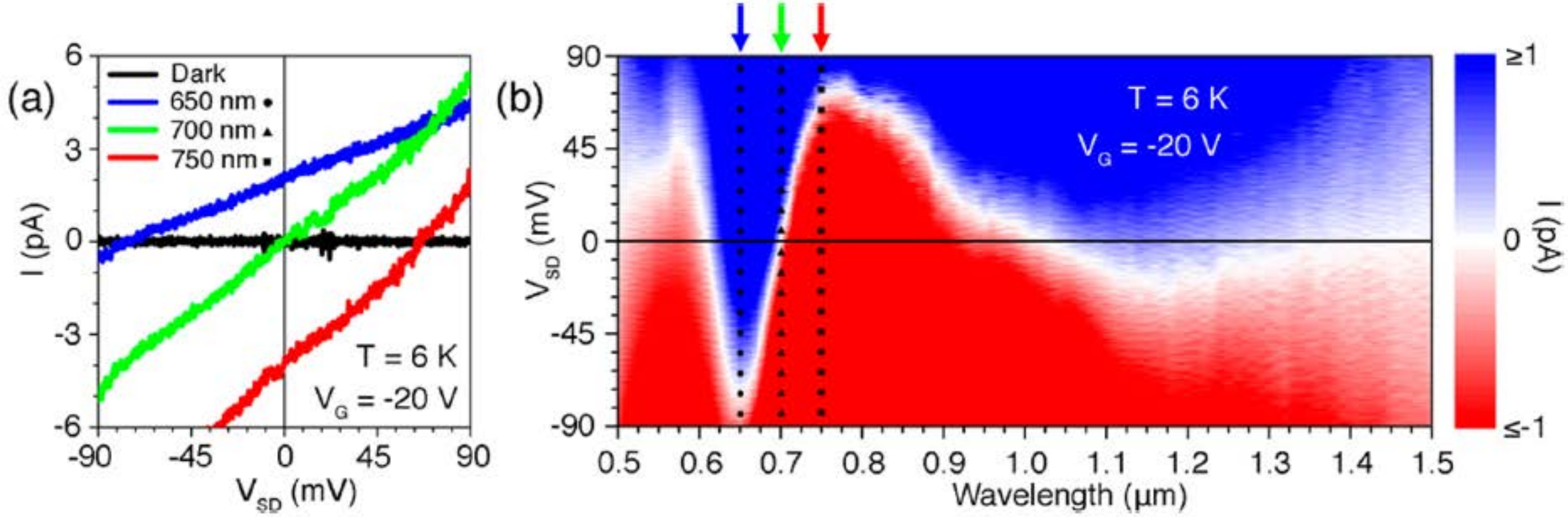




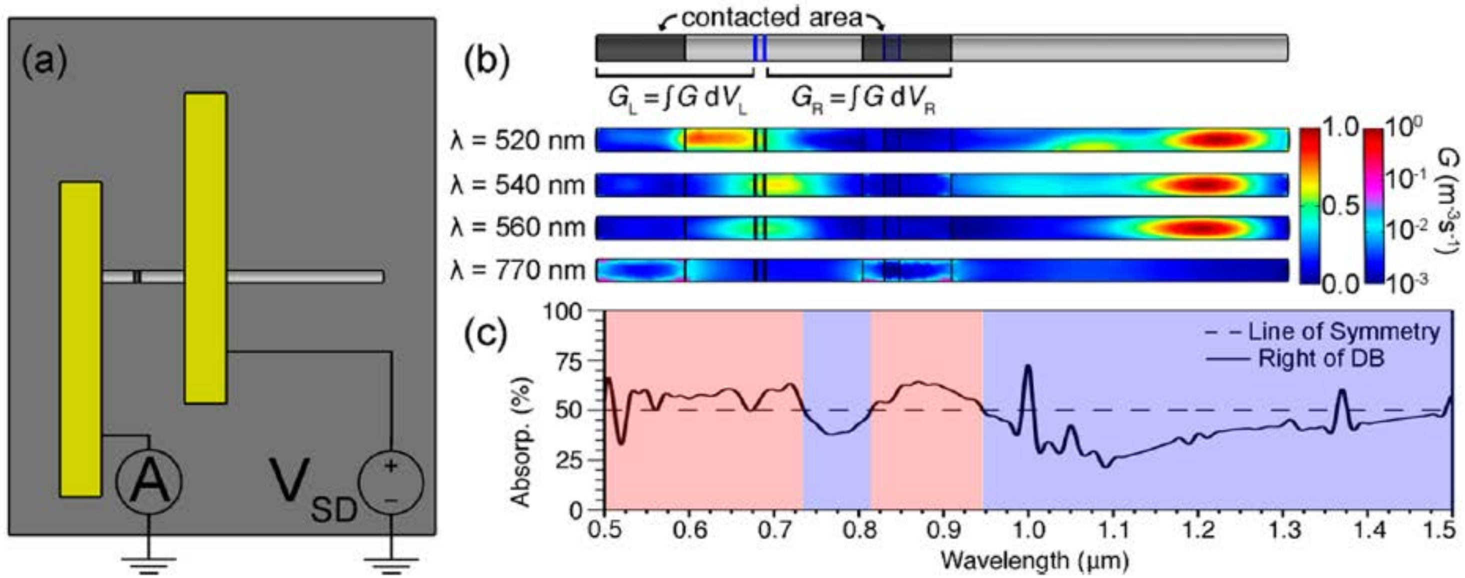
CBE grown InAs/InP/InAs nanowire

Wavelength-sensitivity (Double-barrier device)

Experiment



Model

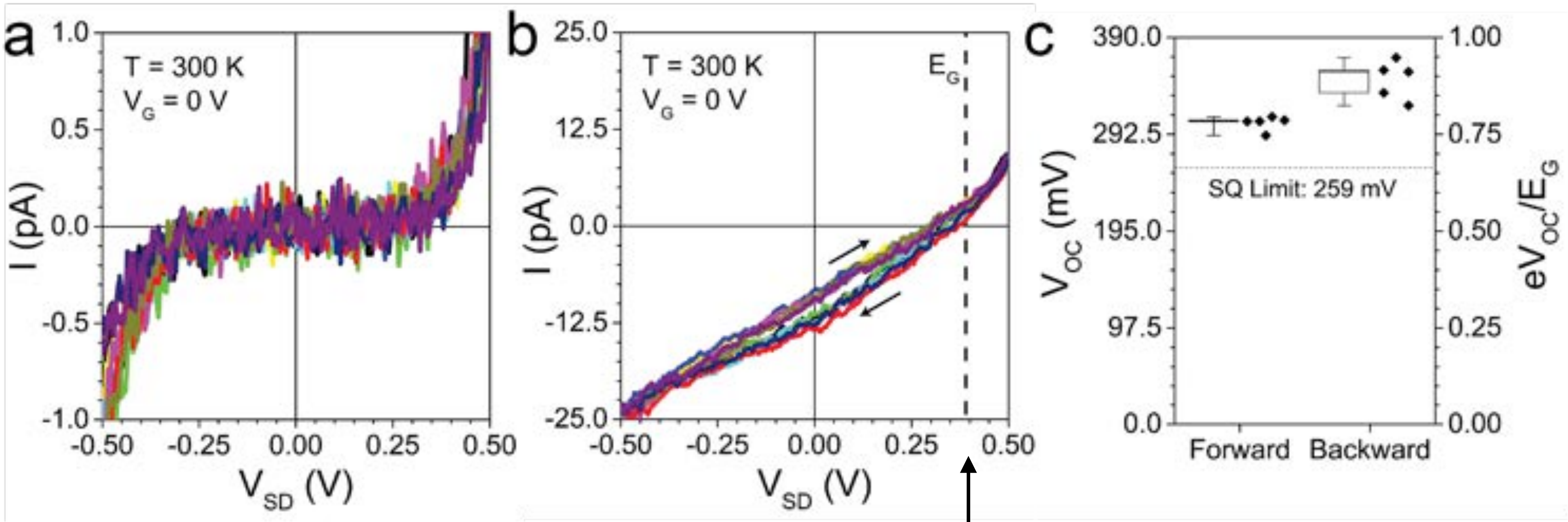


S. Limpert et al, Nano Lett. **17**, 4055 (2017)

Photovoltaic power production (without pn-junction!)

Single-barrier (thermionic) device

V_{OC} > 90% of the bandgap



E_g of WZ InAs
≈ 0.39 eV

S. Limpert et al, Nano Lett. **17**, 4055 (2017)

S. Limpert et al., Nanotechnology **28**, 43 (2017)

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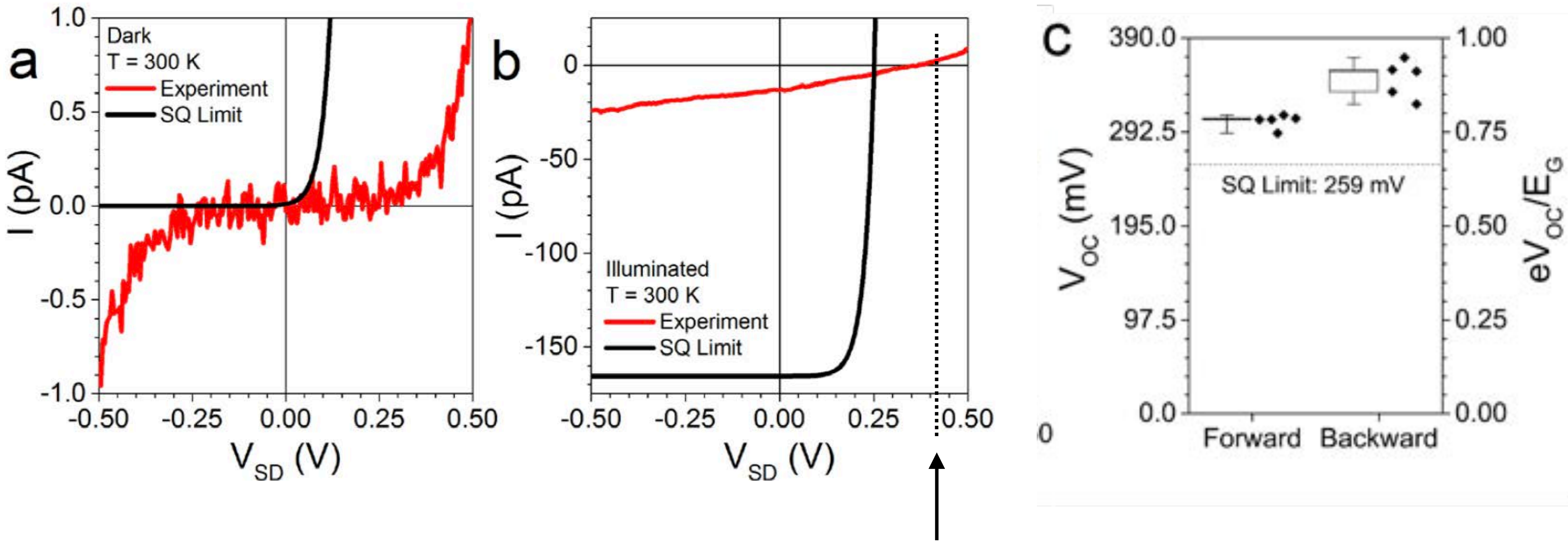
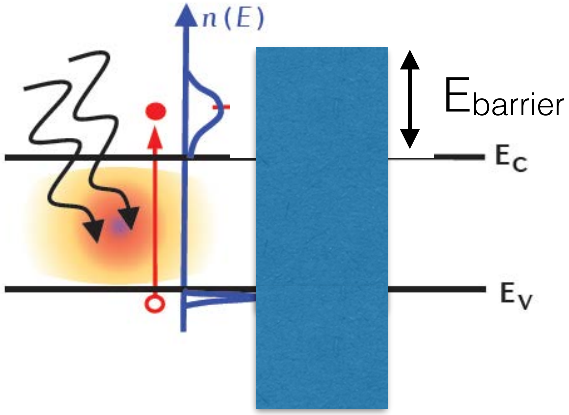


E_g of WZ InAs ≈ 0.39 eV

S. Limpert et al, Nano Lett. **17**, 4055 (2017)

S. Limpert et al., Nanotechnology **28**, 43 (2017)

Thermionic interpretation



Thermionic interpretation:

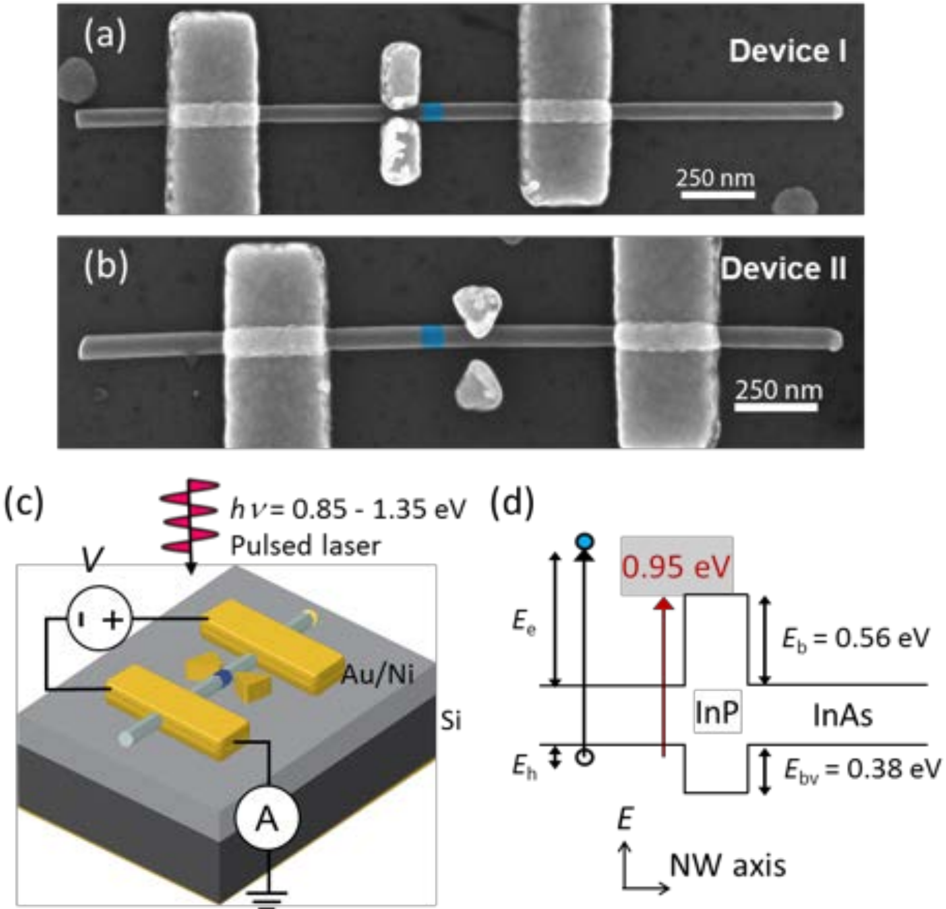
$V_{oc} = (k/e)\ (2 + E_{barrier}/kT)\ \Delta T_{carrier}$

$V_{oc} \approx 0.35$ V is consistent with $\Delta T_{carrier} \approx 170$ K

S. Limpert et al, Nano Lett. **17**, 4055 (2017)

Since ΔT in this interpretation is the carrier temperature, phonon-mediated heat flow is irrelevant to the efficiency analysis.

Controlling the light-absorption hot spot



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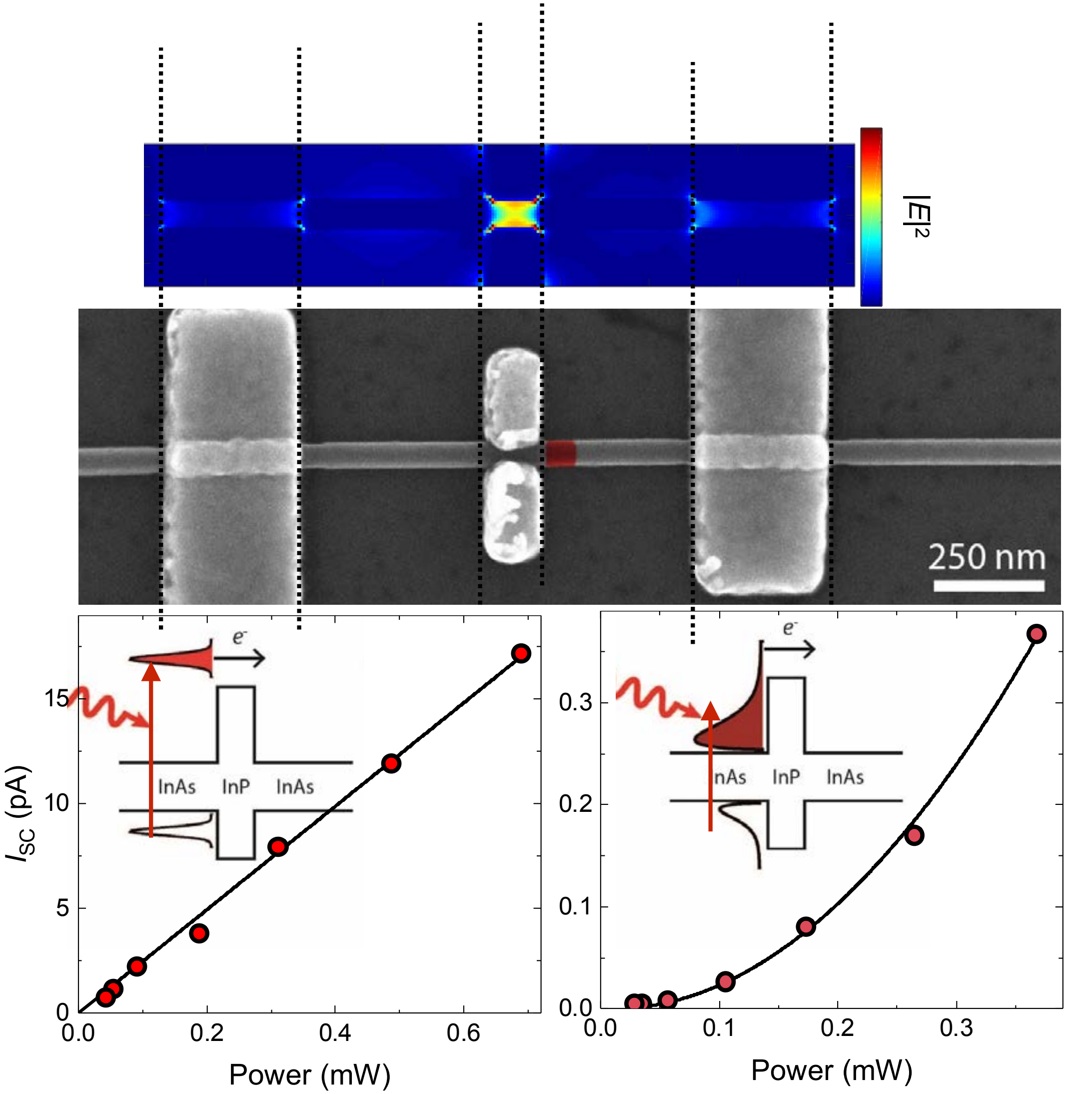
Evidence of quasi-ballistic extraction of hot carriers



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