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NOVEL THEORIES FOR THE THERMOELECTRIC EFFECT

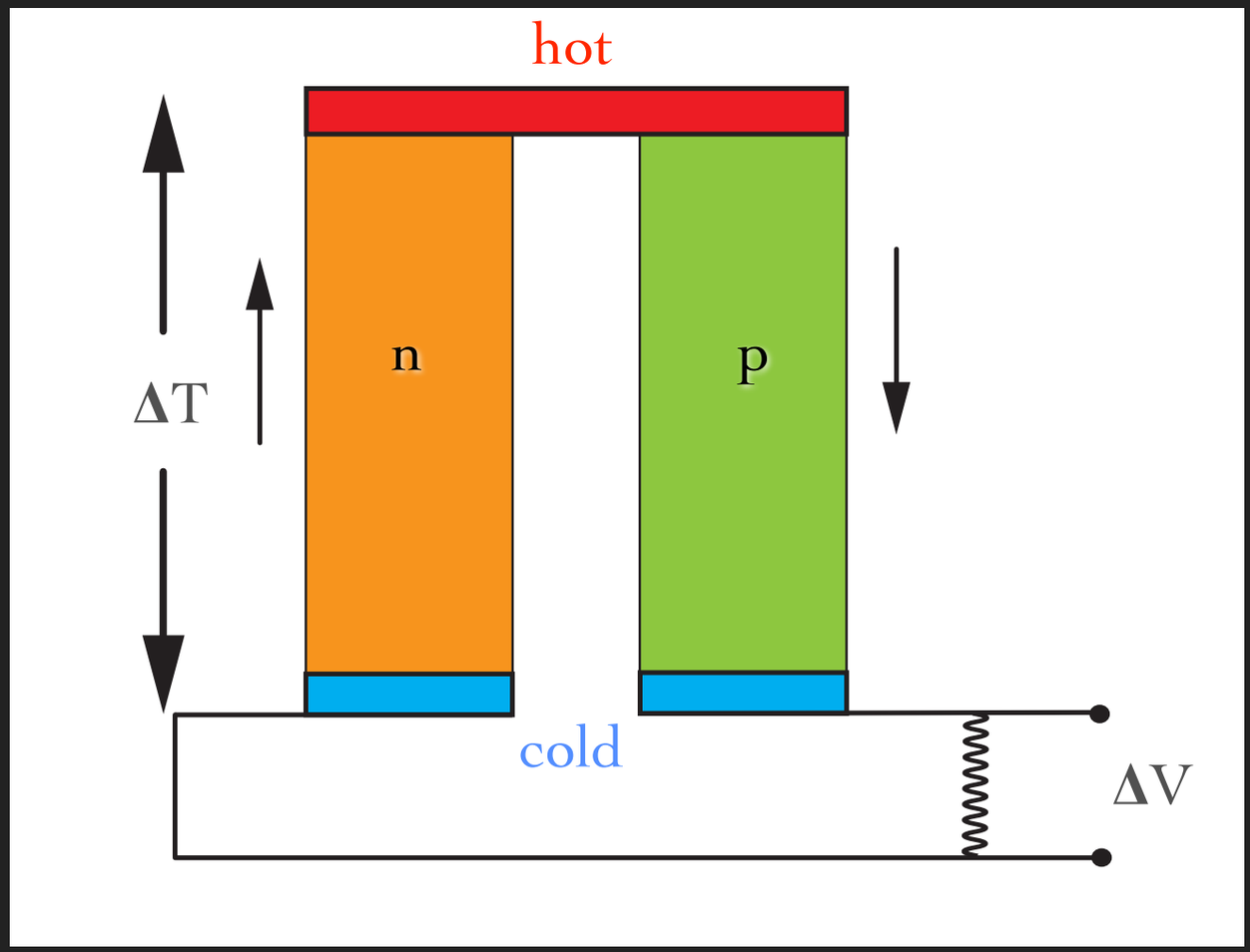
NanoSpain 2017 - Donostia-San Sebastian

INTRODUCTION

- Thermoelectricity is the ability of a device to convert heat into electrical current
- Applications are many and potential game-changer especially when combined with other technologies
- Device have potential long-life time since there are not moving parts
- Actual limit is the efficiency still too low for large scale applications

A SIMPLIFIED SCHEMATICS

- The junctions of a two semi-conductor ring are kept at different temperatures
- Electrons and holes diffuse from hot to cold bath generating an electrical current
- Efficiency is calculated as the power delivered to the load with respect to the heat absorbed
- Generally the efficiency is expressed in terms of the figure-of-merit *ZT*



THERMOELECTRIC EFFICIENCY

Seebeck coeff.

electrical conductance

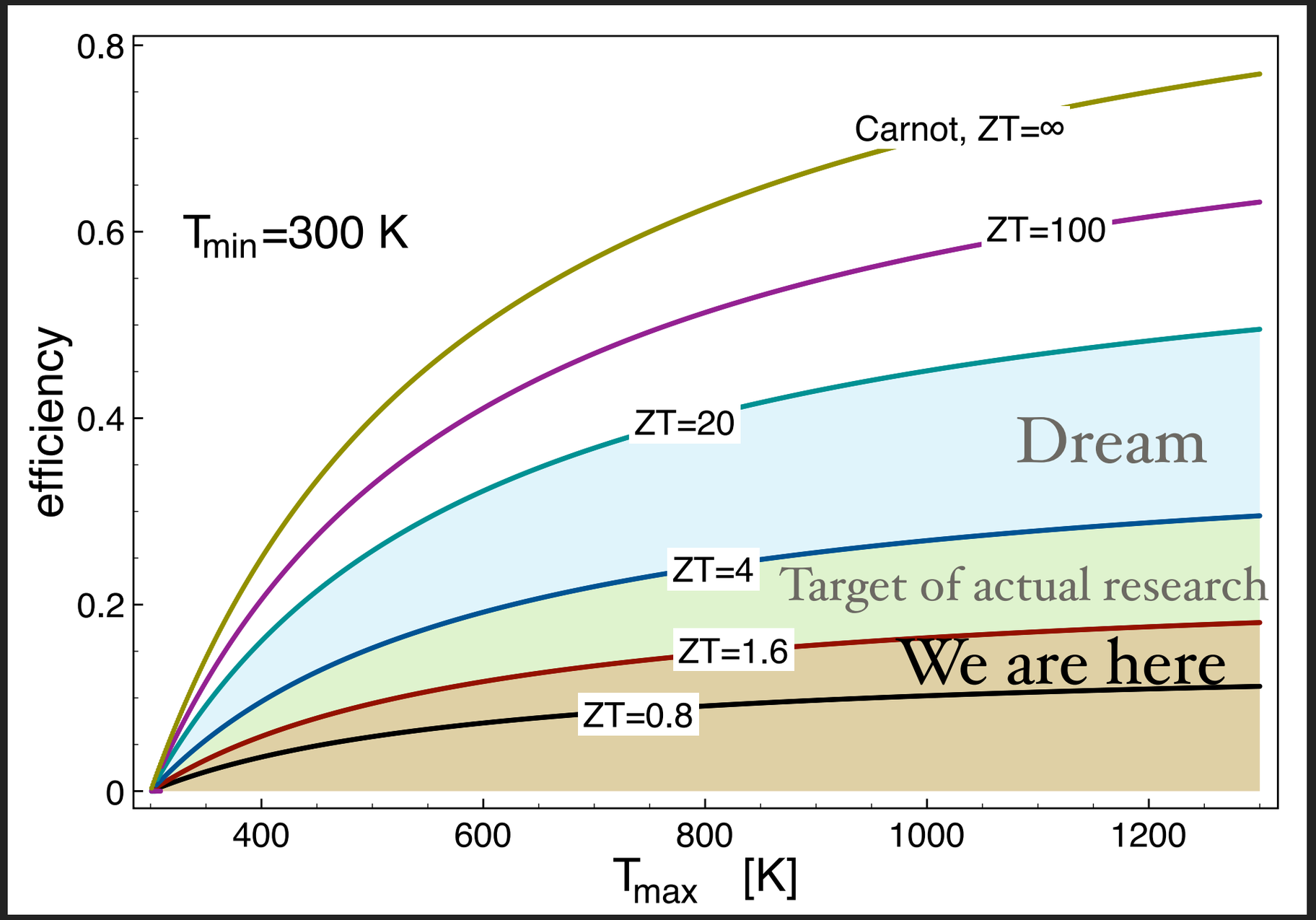
$$ZT = \frac{S^2\sigma}{\kappa}T = \frac{S^2\sigma}{\kappa_L + \kappa_e}T$$

thermal conductance

working temperature

$$S = -\frac{\Delta V}{\Delta T}$$

A FIGURE IS WORTH THOUSAND...



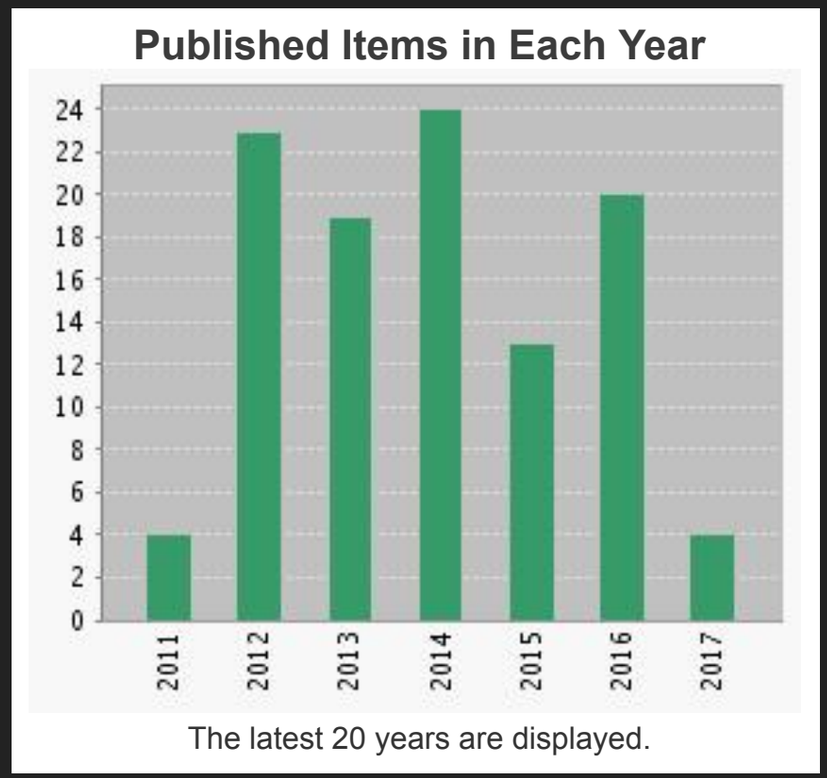
Optimising ZT is a difficult task since the transport coefficients are usually **correlated**

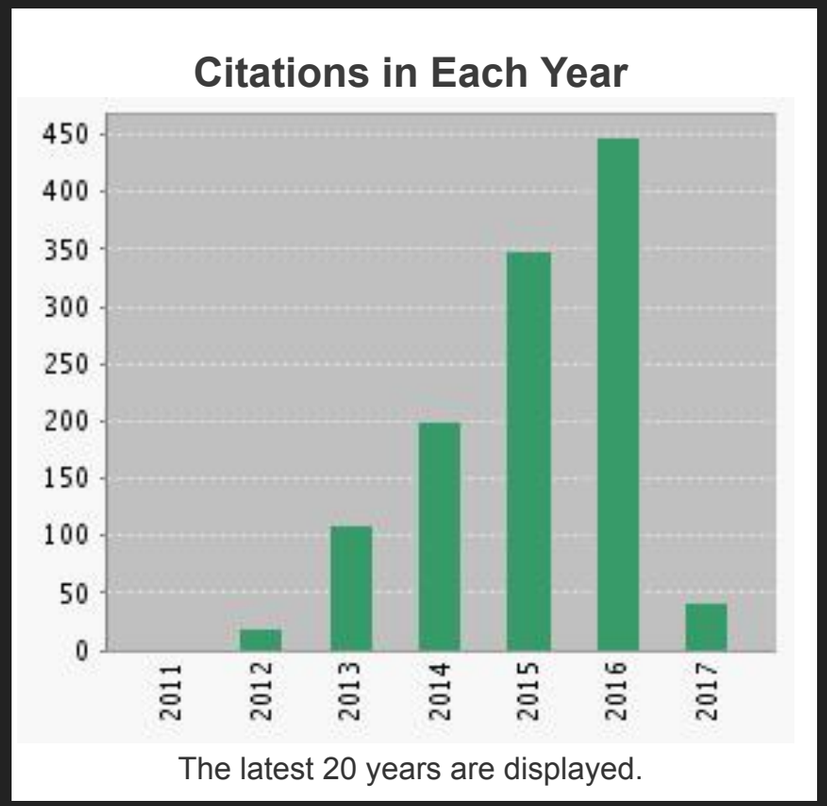
THE NANO-THERM PROJECT

- Thermoelectricity is considered a technological priority by the Spanish government
- In 2010, a CONSOLIDER project coordinated by C. S. Sotomayor-Torres was funded to bring Spain at the forefront of the international thermoelectric community
- The NanoTHERM project developed over 6 years and in this time obtained state-of-the-art and "world record" results
- A review of the overall progress has been recently published in O. Caballero and R. D'Agosta, *ECS J. Solid State Sci. Technol.* **6**, N3065-N3079 (2017).

SOME STATISTICS (SO FAR)

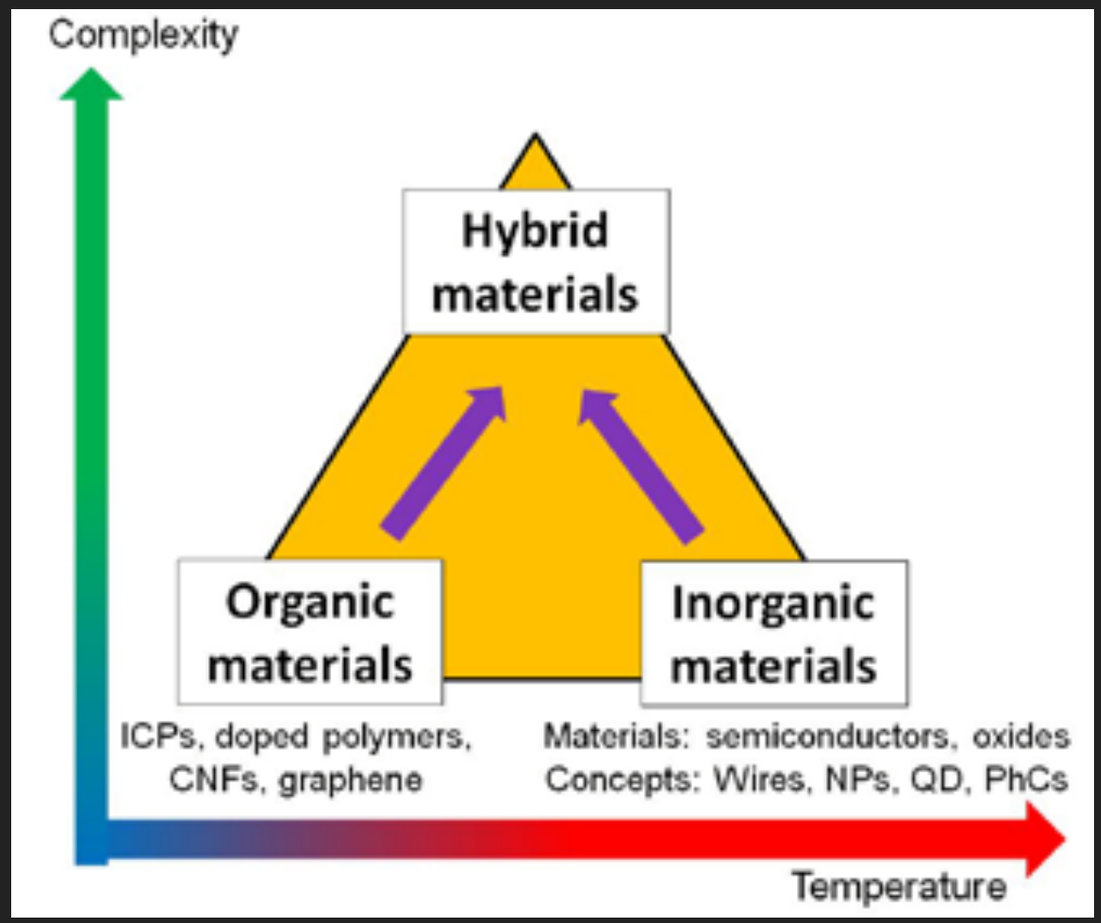
- 9 groups from across Spain + 1 SME
- 22 scientists (PhD students, technicians, postdocs) have been paid directly by the project
- 1 national patent
- More difficult is to estimate the future impact of this project (but members are winning prizes for their start-up…)





OUR APPROACH

- We developed a synergy between theory and experiments
- We rationally expanded the field of both organic and inorganic thermoelectric materials
- We rationally designed hybrid materials able to perform better than the single components.



For more details about the experimental progresses made in this field, please attend the Friday talks by

Dr. Olga Caballero-Calero and Dr. Marisol Martin

SOME (OUTSTANDING) ACHIEVEMENTS

- Built an national network which is now also internationally recognised
- Established some common measurement techniques for the thermoelectric parameters: important for standardisation and comparison
- World record for the smallest BiTe nano-wires (below 12 nm)! (See Olga and Marisol's talks)
- Manyfold (orders of magnitude) increase of the efficiency in organic thermoelectric devices
- Novel theoretical approaches to go beyond the actual state-of-the-art

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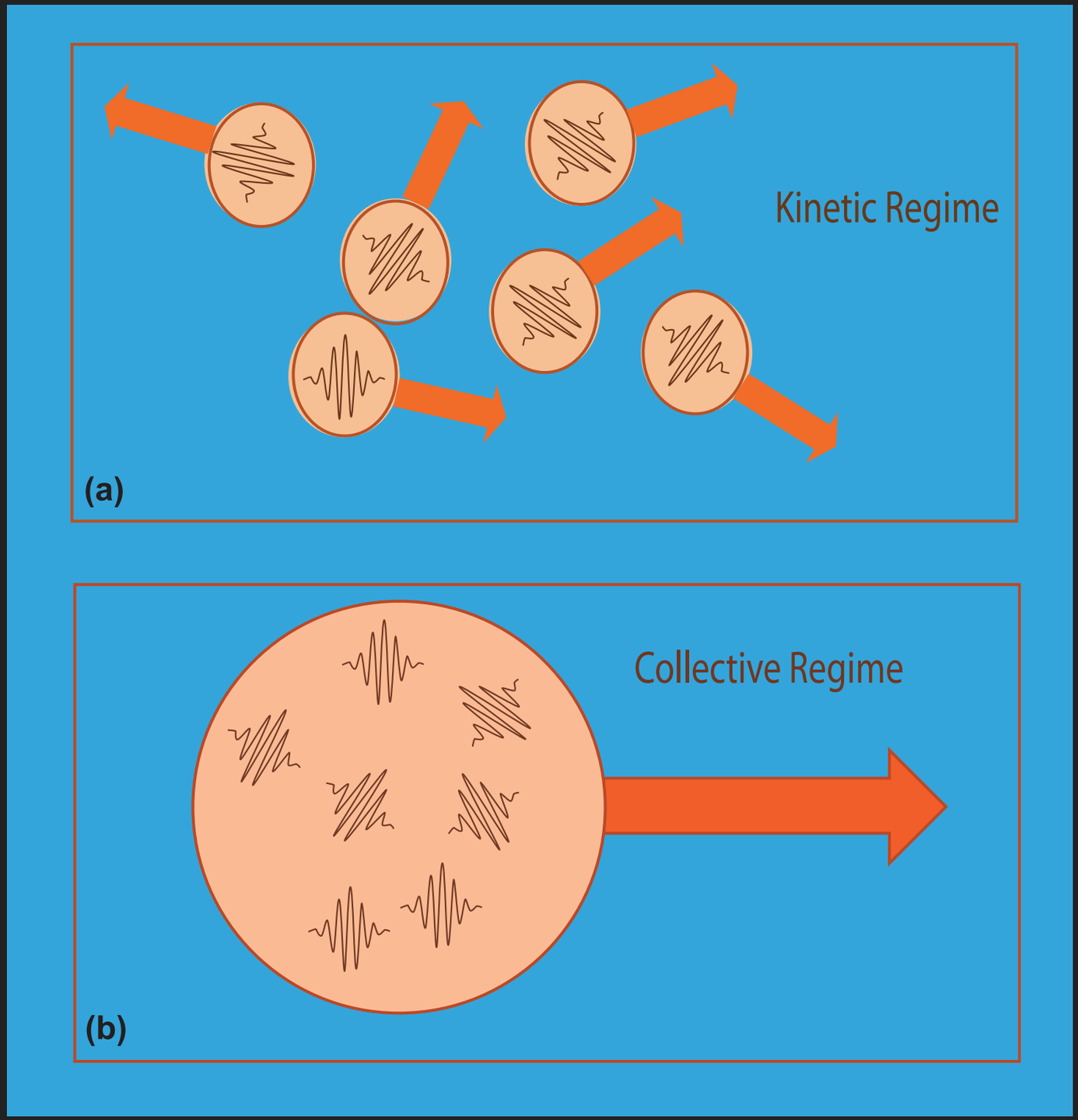
Now, let me shift gear…

KINETIC-COLLECTIVE MODEL

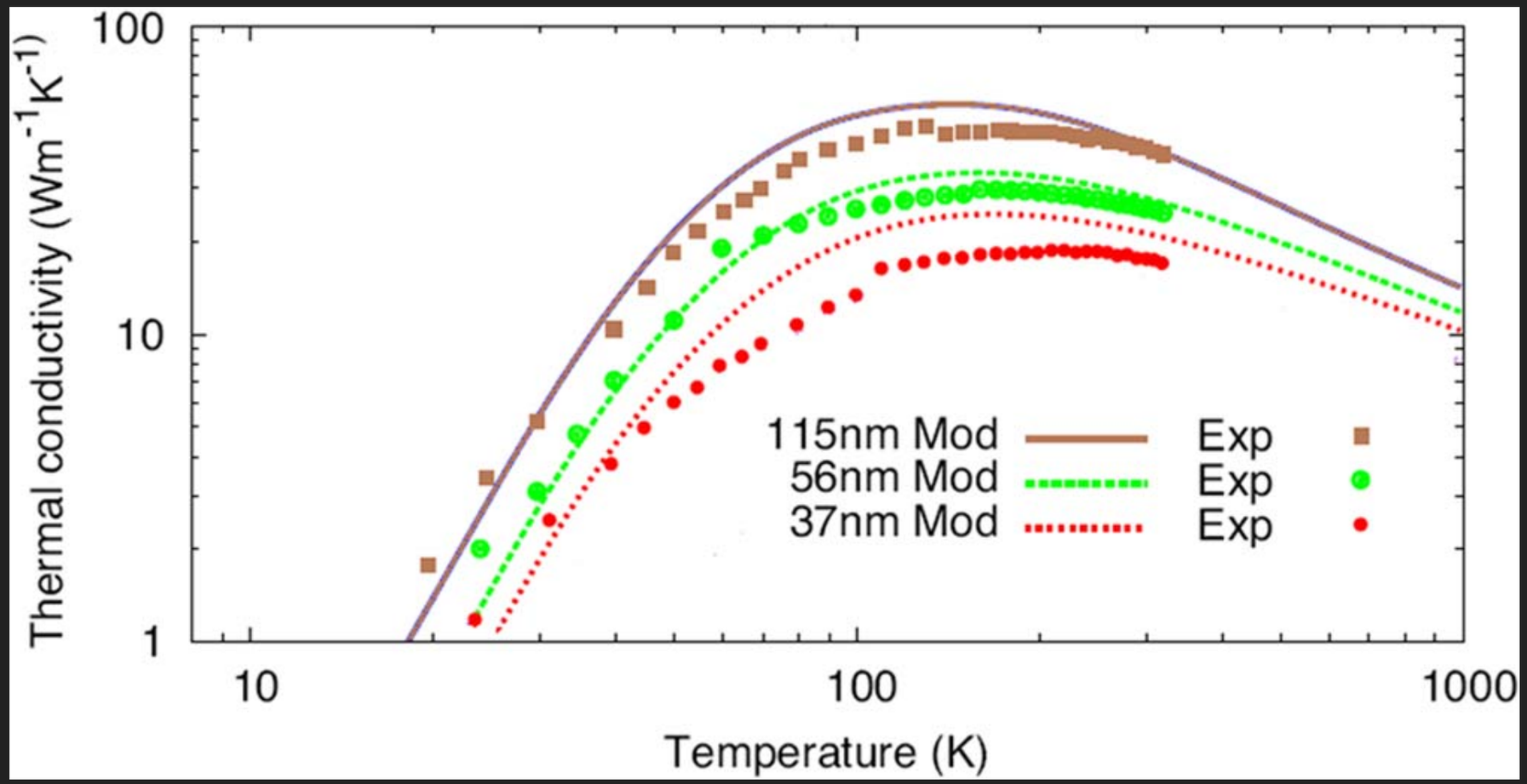
- Describe phonon thermal transport in a Boltzmann transport formalisms
- Separate two time-scales: a kinetic and a collective regime
- The collective regime is triggered by normal collisions
- Novel particle sometimes dubbed "relaxon"

C. de Tomas, A. Cantarero, A. Lopeandia, and F. Alvarez, Journal of Applied Physics, 115 (2014).



SILICON NANOWIRE THERMAL CONDUCTANCE

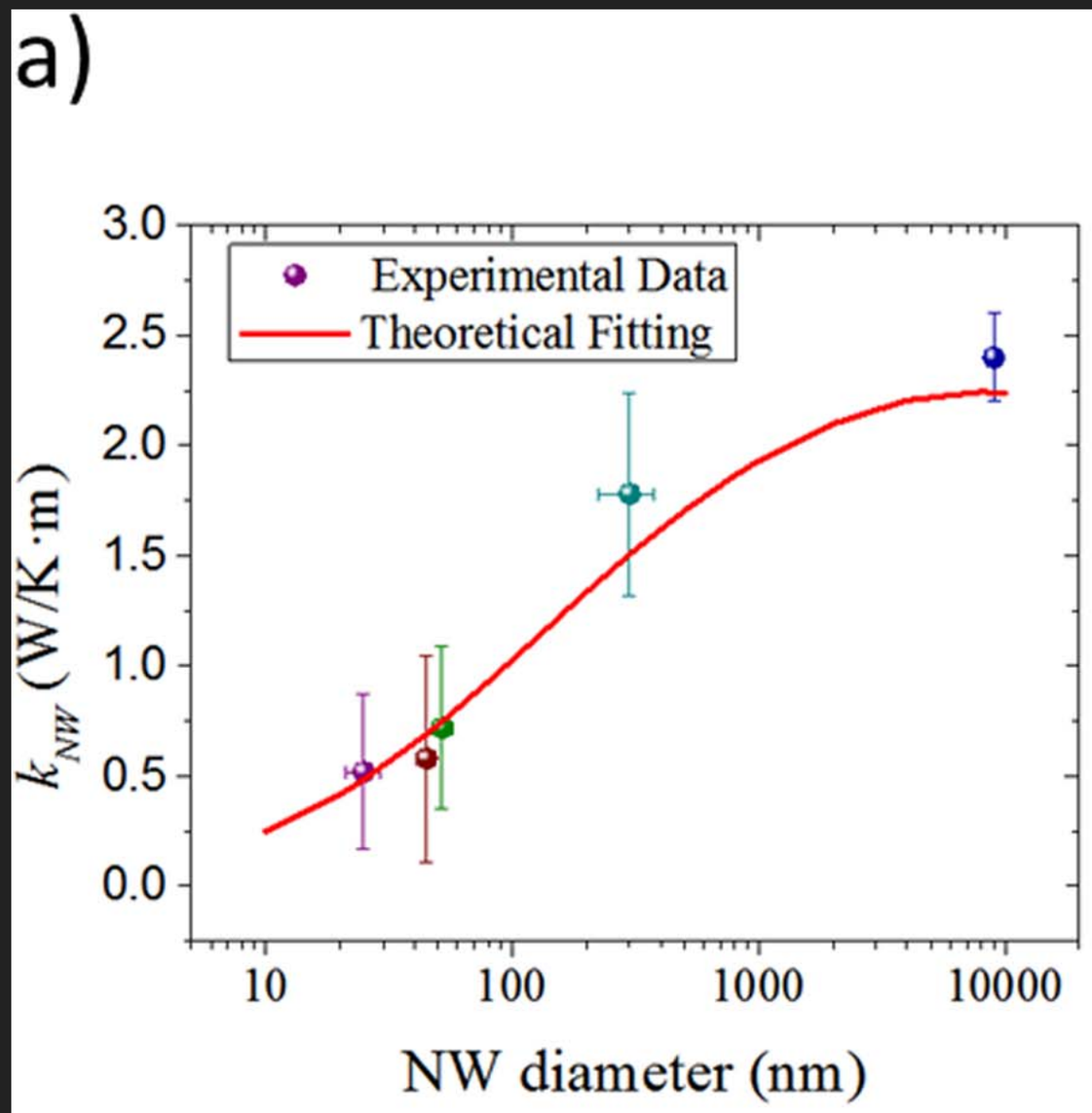
- The kinetic-collective model is used to study thermal transport in Si nano wires
- It captures quite well the behaviour over a range of diameters down to ten of nano-meters
- Physical parameters are taken from the bulk Si!

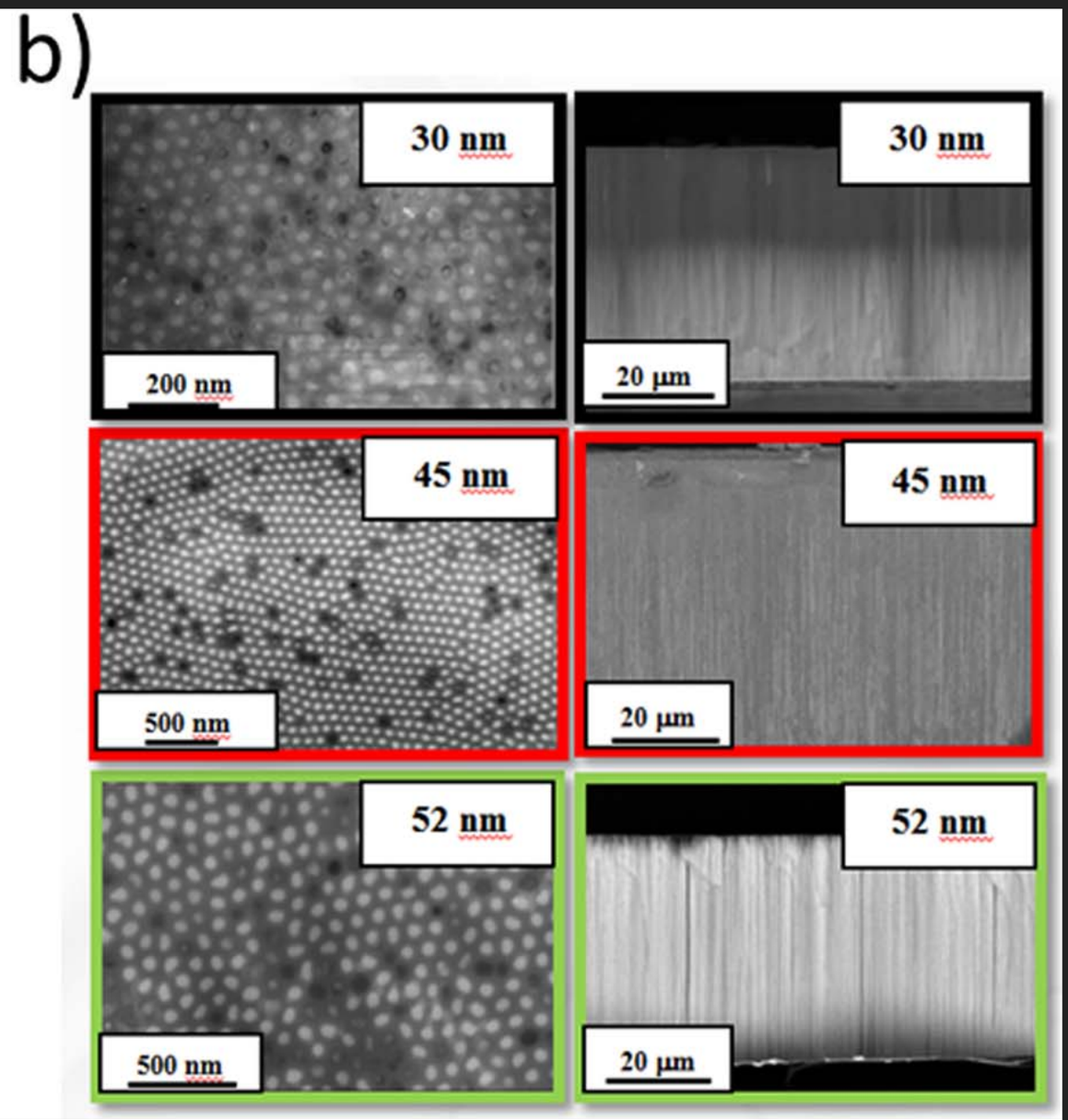


BISMUTH-TELLURIDE

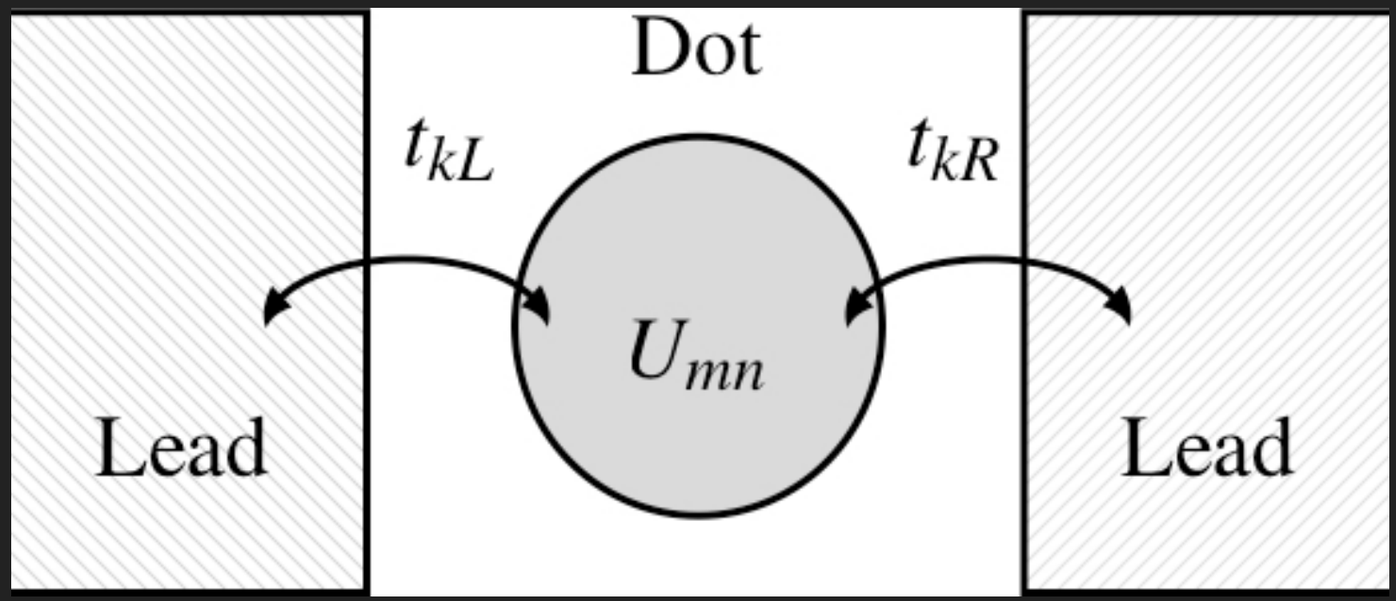
O. Caballero-Calero, et al. Materials Today: Proceedings, 2, 620 (2015).

- The kinetic-collective model can be applied to other nano-wires reaching excellent agreement with experiments.





STRONG ELECTRON CORRELATION EFFECT ON TRANSPORT



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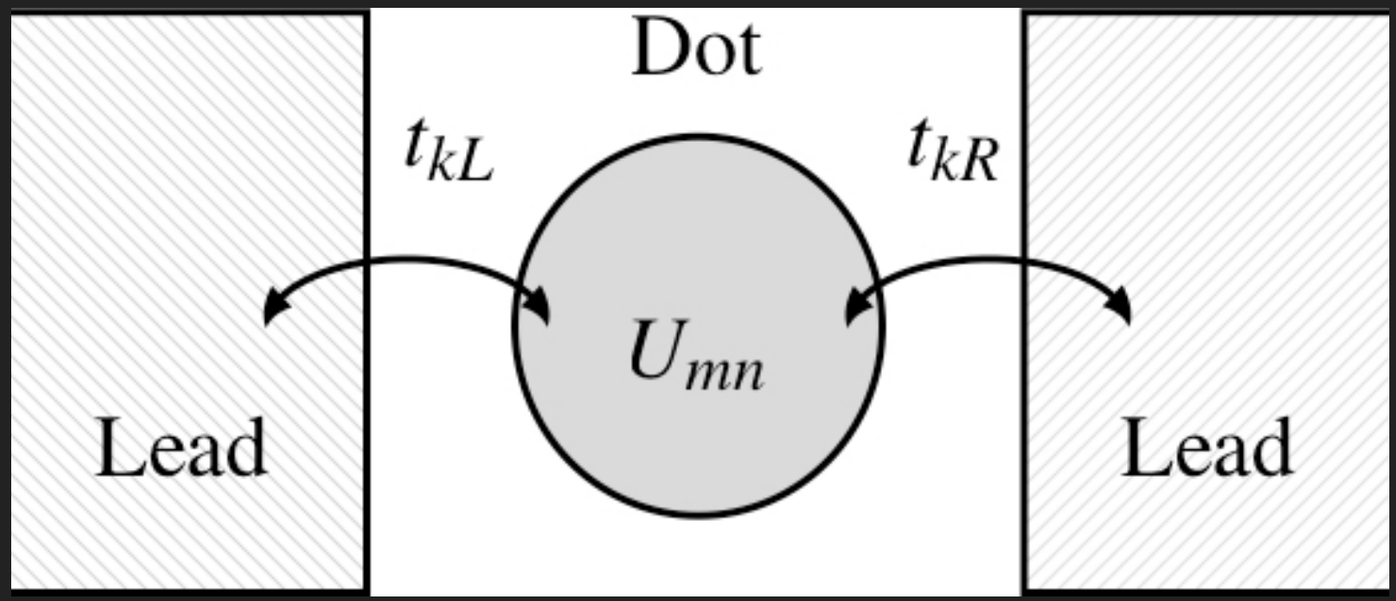

How does Density functional theory works for this system?

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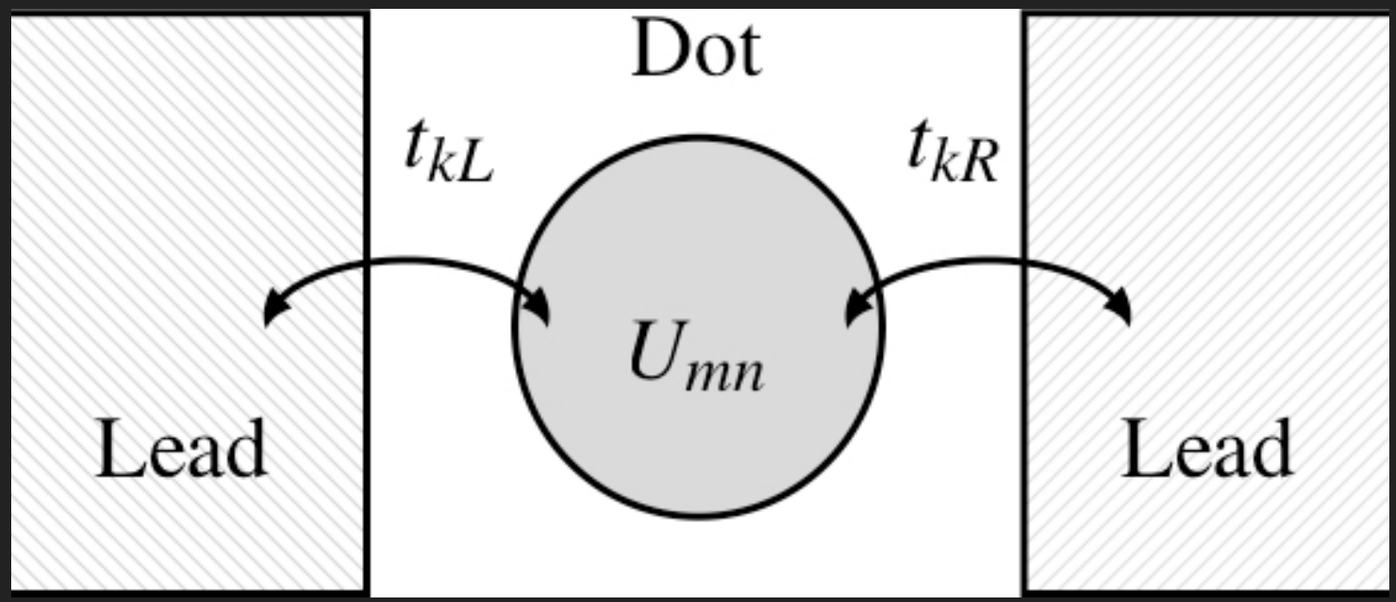

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Badly!

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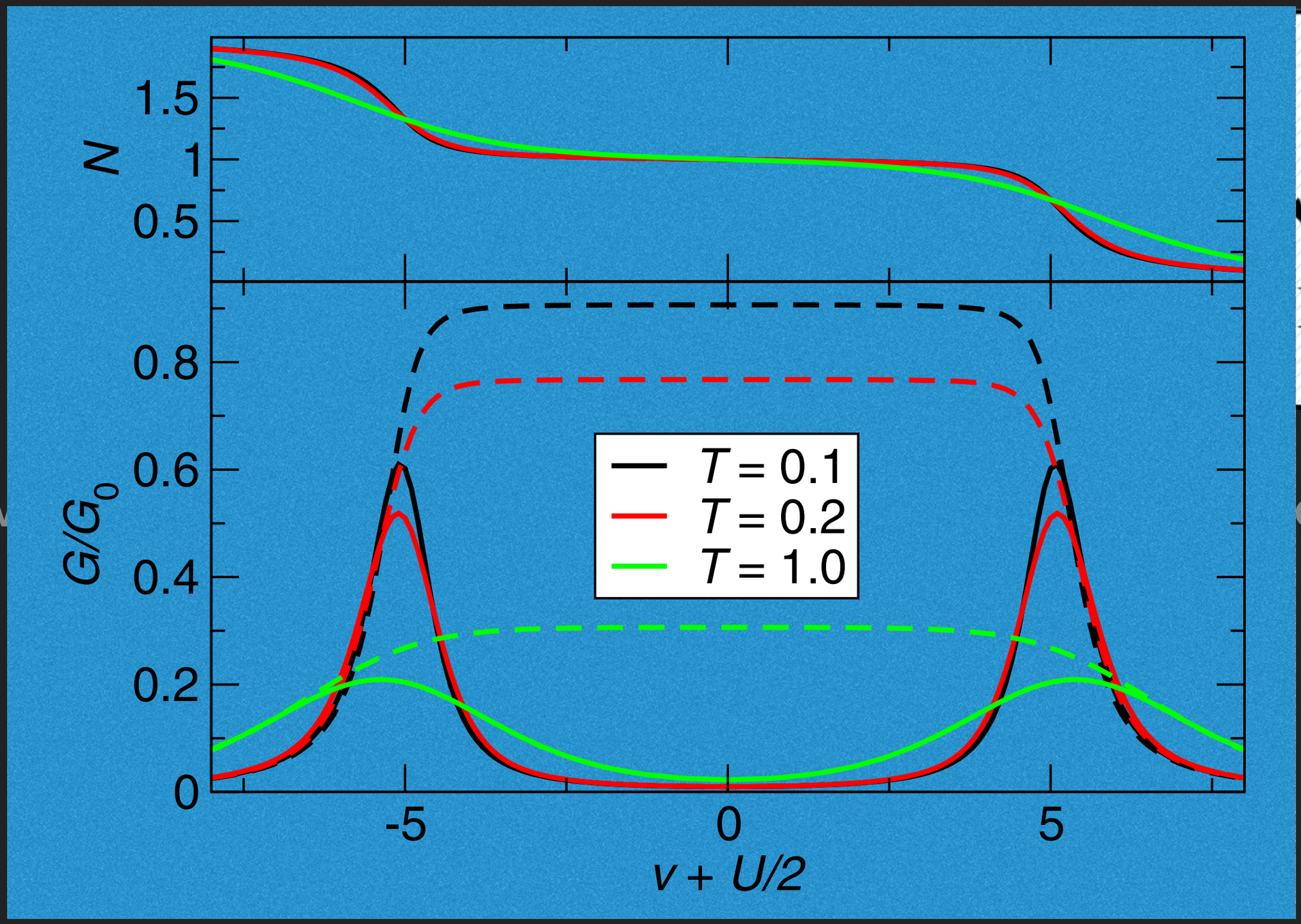


Lead

How em?

DYNAMICAL CORRECTIONS

Remember: DFT entails all exchange and correlation effects in the V_{Hxc} potential

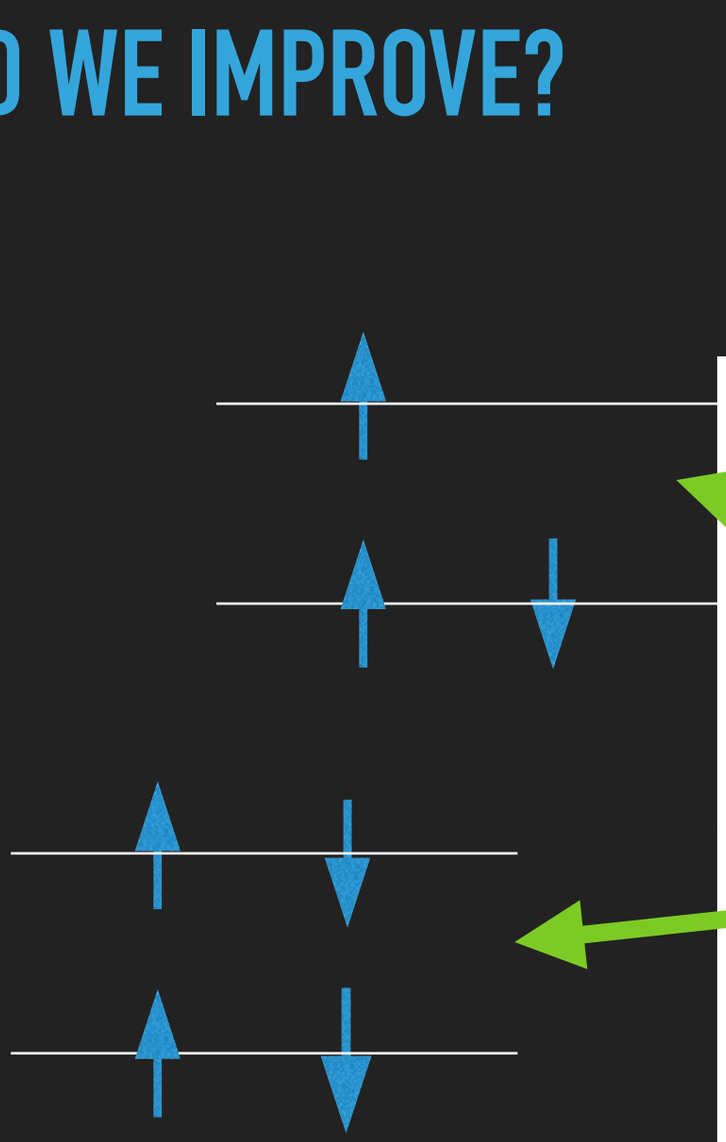
$$G/G_{KS} = \frac{1+R}{1+\frac{t}{t_L t_R} G_{KS} \frac{\partial V_{Hxc}}{\partial N}}$$

$$S = S_{KS} + \boxed{\frac{\partial V_{Hxc}}{\partial T}} = \frac{\partial J_{KS}/\partial T}{G_{KS}} + \frac{\partial V_{Hxc}}{\partial T}$$

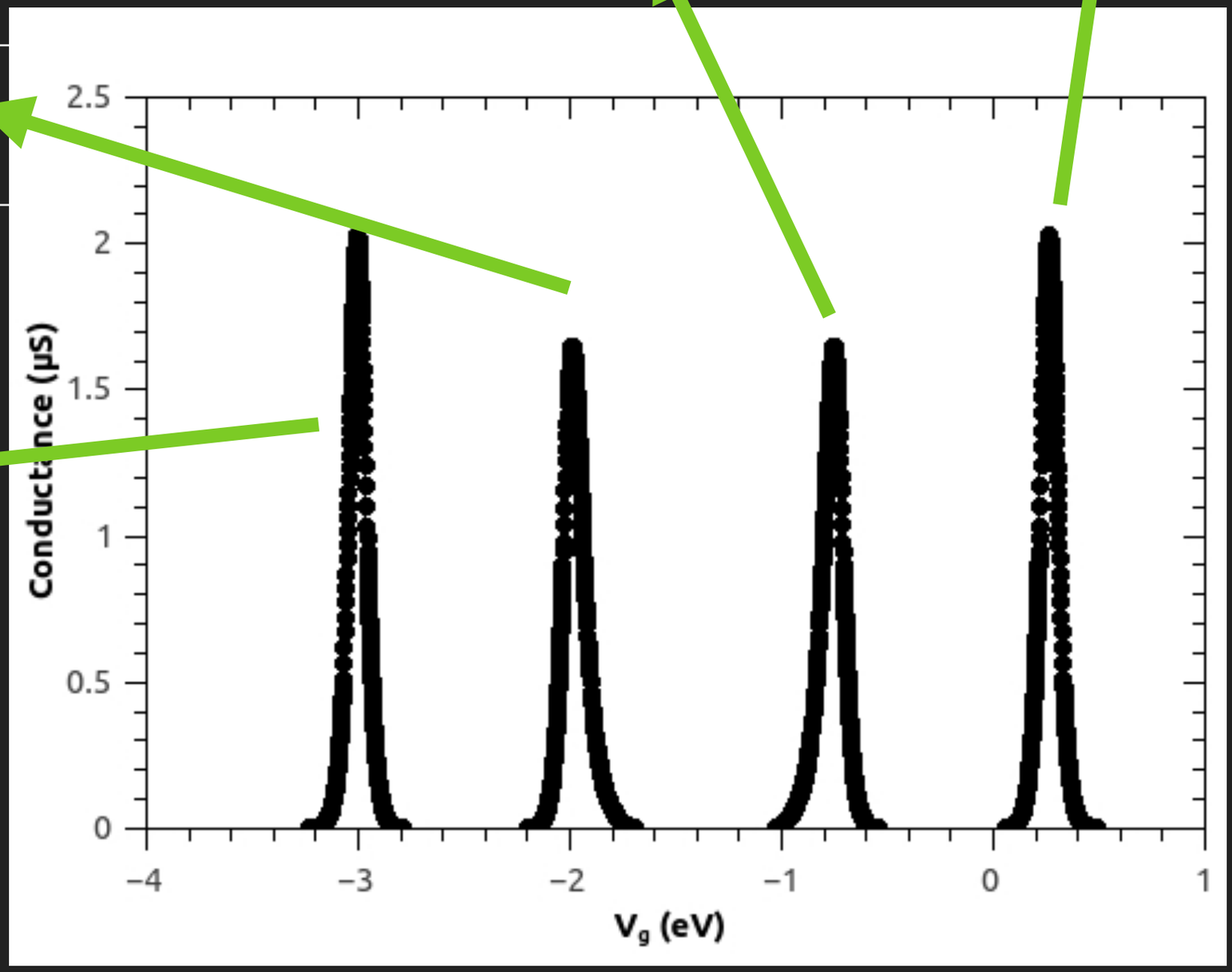
K. Yang, E. Perfetto, S. Kurth, G. Stefanucci, and R. D'Agosta, Phys. Rev. B 94, 081410(R) (2016)

G. Stefanucci and S. Kurth, Nano Letters 15, 8020 (2015)

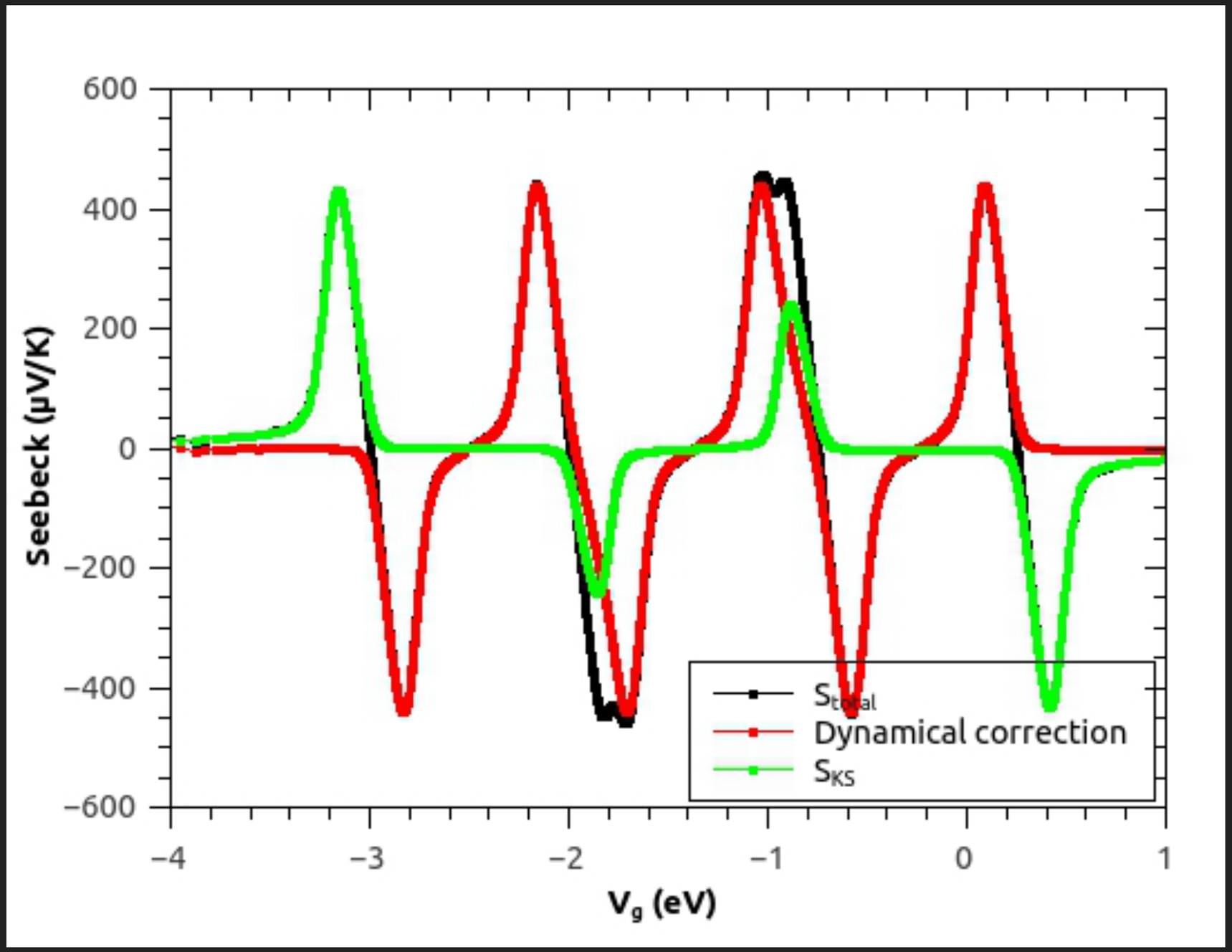
DID WE IMPROVE?



Addition energy = 1 eV



AND THE SEEBECK COEFFICIENT?



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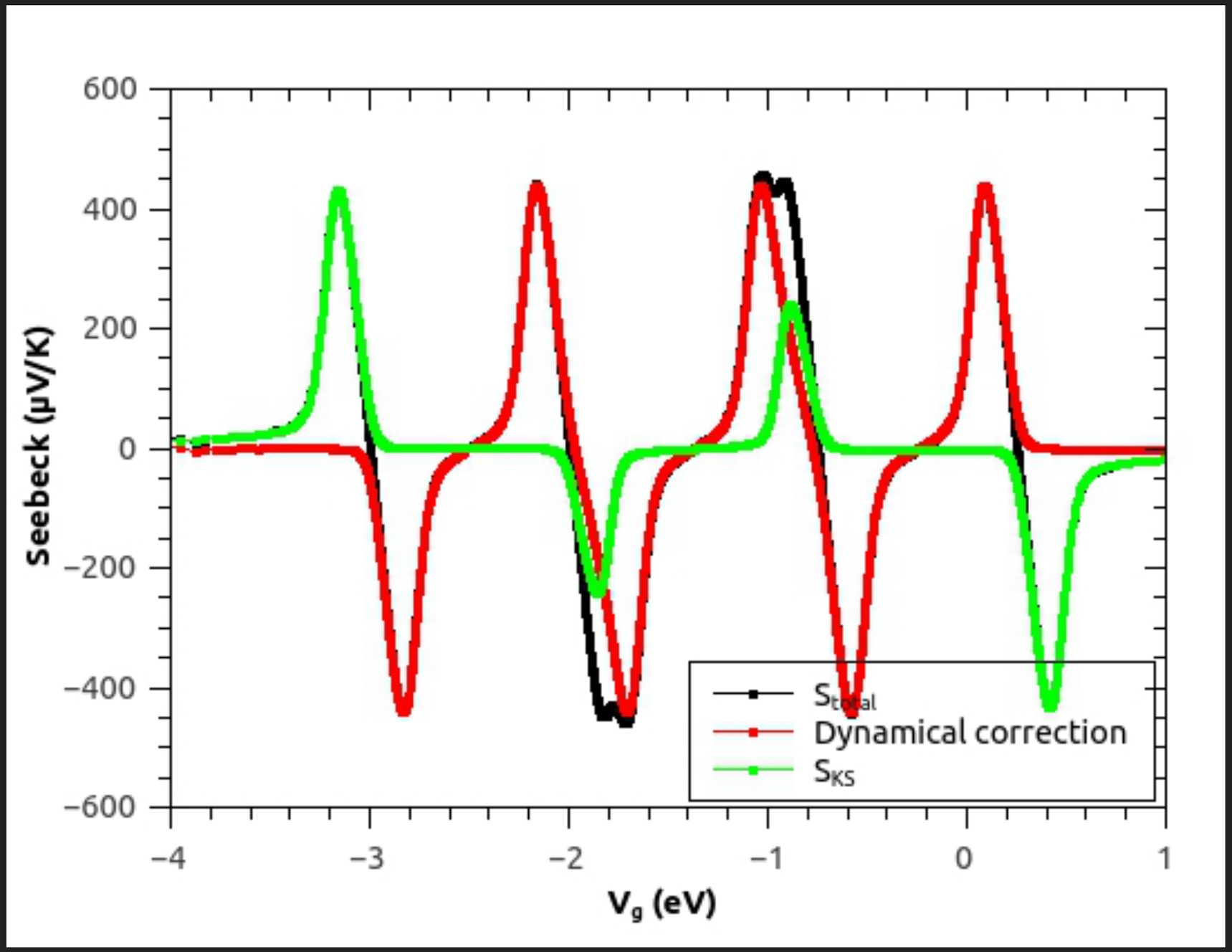
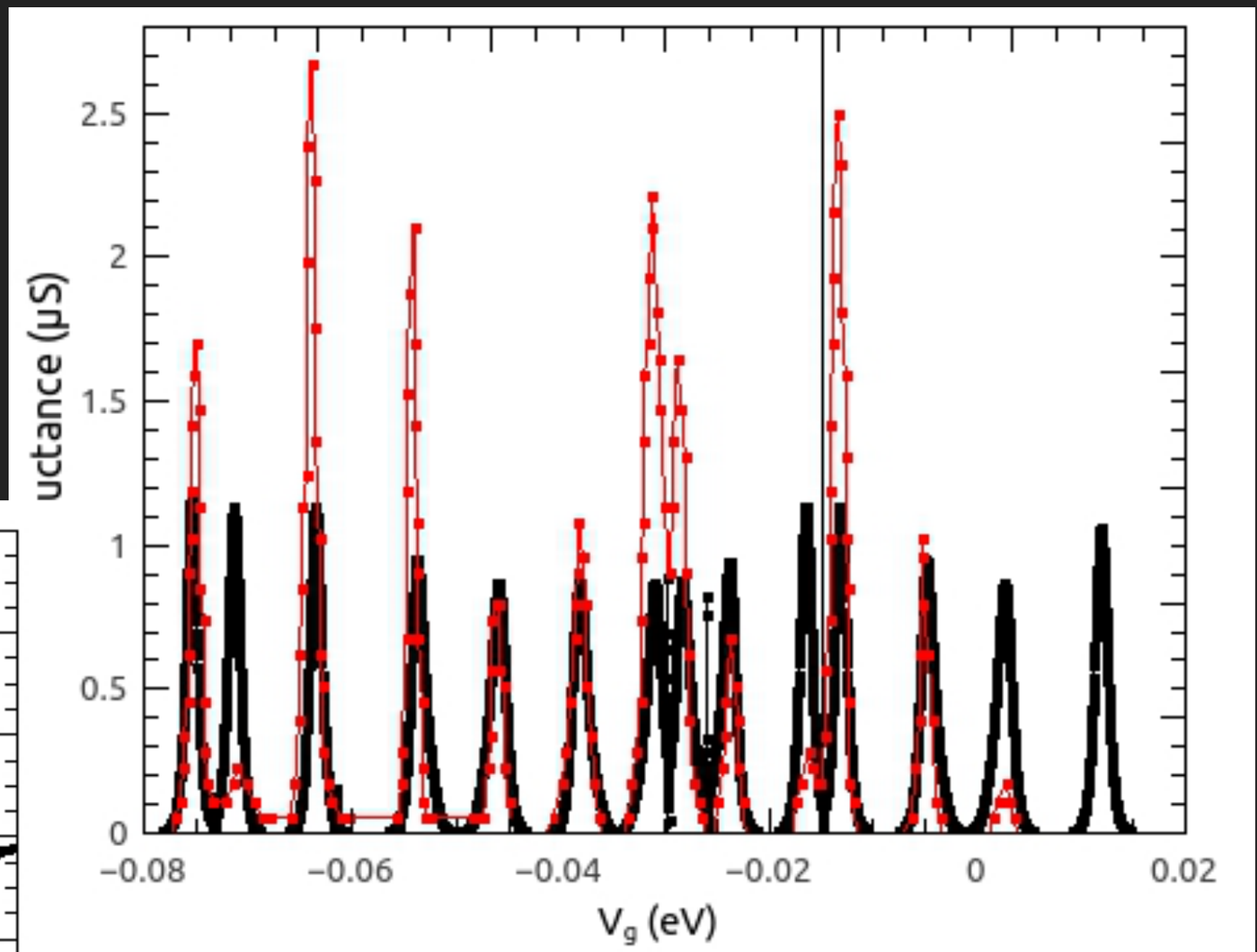
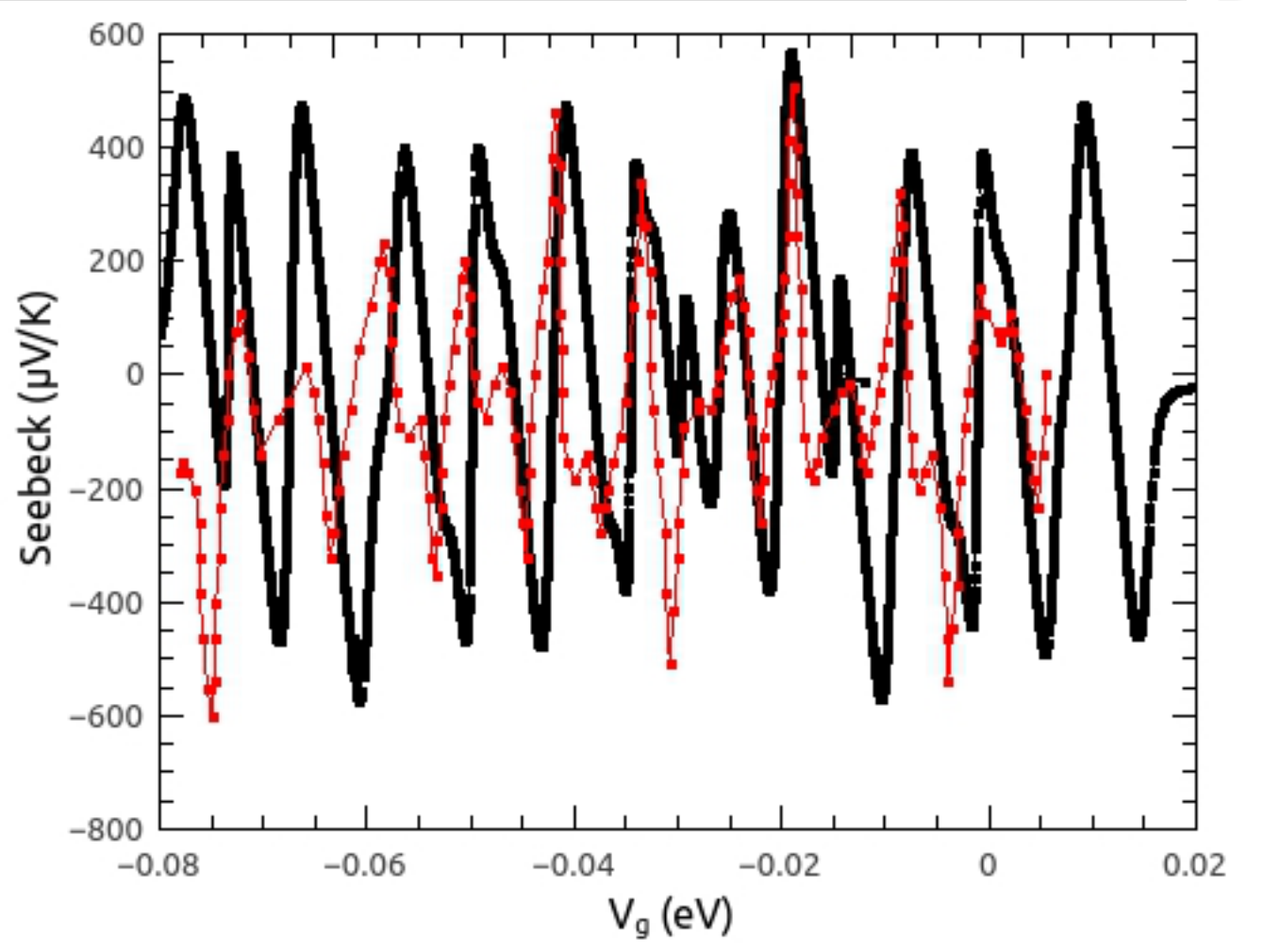


This correction shows the limit of the current approaches

A REAL SYSTEM: CARBON NANOTUBE

EXP: J. P. Small, K. M. Perez, and P. Kim,
Phys. Rev. Lett. 91, 256801 (2003).





THEORY: K. Yang, E. Perfetto, S. Kurth, G. Stefanucci,
and R. D'Agosta, Phys. Rev. B 94, 081410(R) (2016)

A NEW "RED TEMATICA"

- MINECO recently funded our "red tematica" on thermoelectric theory
- IDEA: bring together different theoretical groups to share their expertise
- It is a "theoretical" network but we welcome input from industry and experimentalists
- It is an "open" project, so if you are interested please contact us (Roberto D'Agosta or Andres Cantarero @ U. of Valencia)

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ACKNOWLEDGMENTS

- Nano Spain Conf for the kind invitation
- CONSOLIDER NanoTHERM (all the people there for the amazing trip)
- IKERBASQUE - Basque Foundation for Science
- Universidad del Pais Vasco

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Basque Foundation for Science

MINISTERIO
DE ECONOMÍA
Y COMPETITIVIDAD

Thank you!