Theoretical Condensed Matter Physics
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Quantum and phonon interference enhanced thermoelectricity

Thermoelectric energy harvesting with single molecules and self-assembled monolayers

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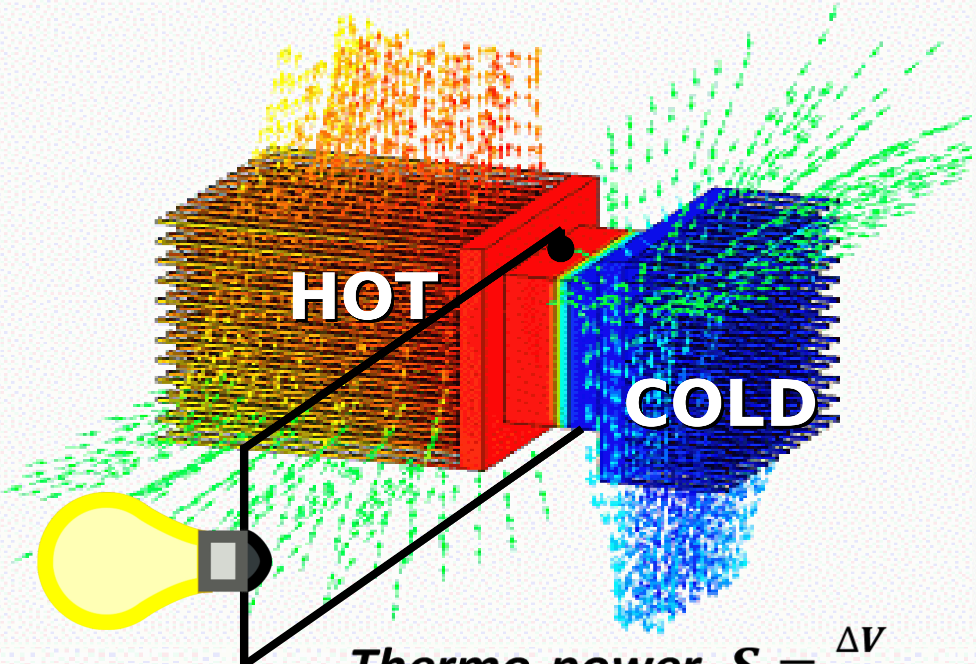
EPSRC Thermoelectric Network UK Meeting,
University of Manchester,
Manchester, UK
www.thermoelectricnetwork.com

Heat conversion to electrical power



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

Thermoelectric figure of merit

$$Z.T = \frac{GS^2}{\kappa_{el} + \kappa_{ph}} \times T$$

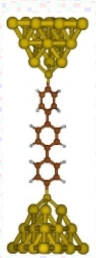
ZT could be infinity!!

G: **Electrical conductance (1/Ω)**
measure of charge flow

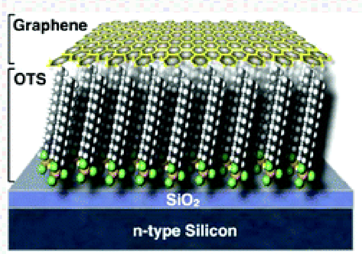
S: **Thermopower (Volt/Kelvin)**
measure of thermoelectric power generation

κ: **Thermal conductance (Watt/Kelvin)**
measure of heat flow due to the electrons (el) and phonons (ph)

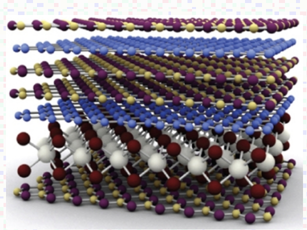
T: **Themperature (Kelvin)**



Single molecules



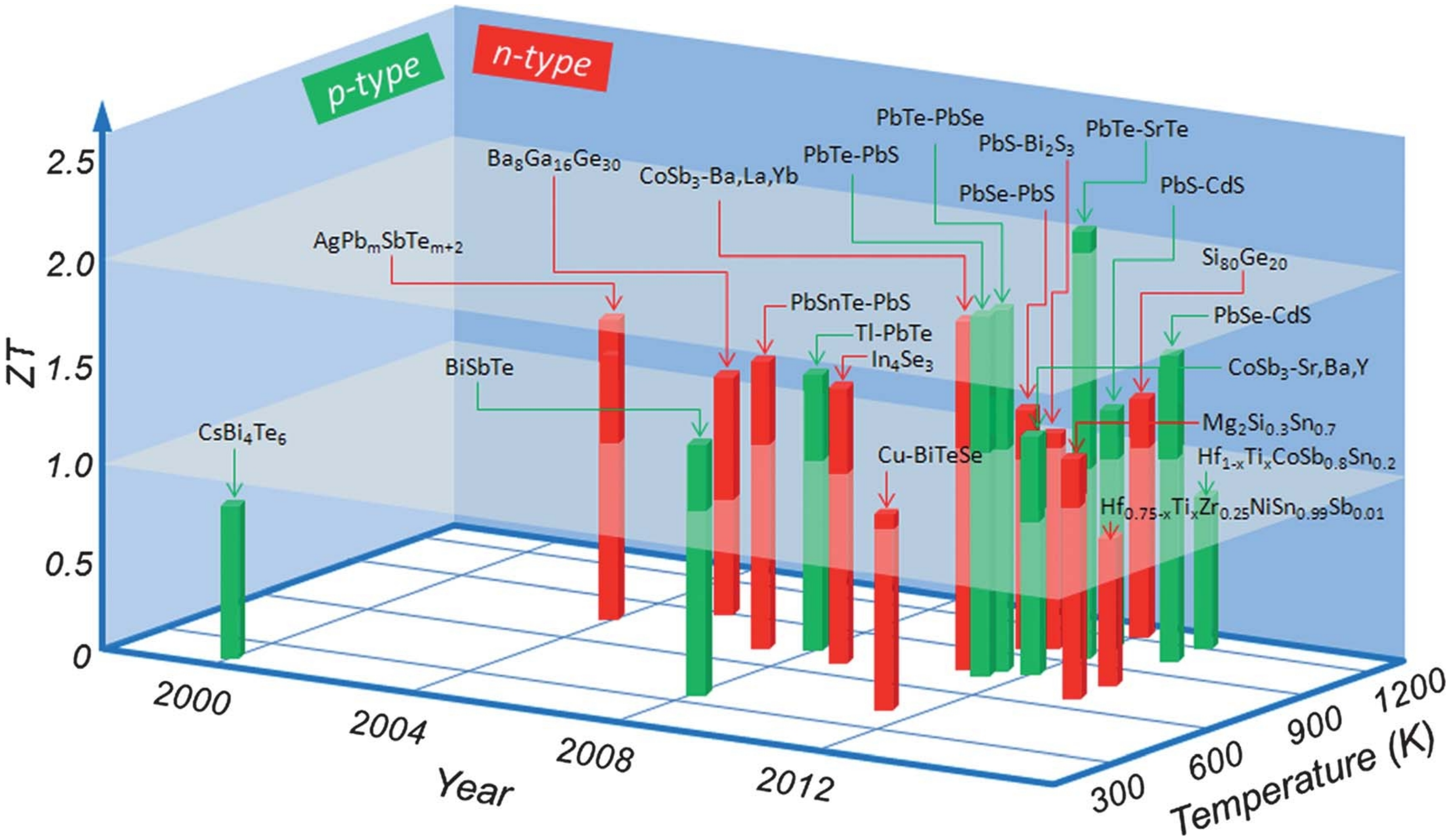
SAMs



vdW heterostructures

State-of-the-Art high ZT materials

The highest value of ZT known so far is ZT = 2.6 ± 0.3 at 923 K in tin selenide (SnSe).



Zhao, et al. Energy Environ. Sci. , 2014, 7 , 251; Zhao, et al. Nature 508.7496 (2014): 373

Why maximising ZT is not easy?

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$$ZT = \frac{GS^2T}{\kappa_{el} + \kappa_{ph}}$$

ZT_{el}

$G\uparrow, S\uparrow, \kappa_{el}\downarrow, \kappa_{ph}\downarrow$

Simultaneous phonon and electron engineering

1 Enhance ZT_{el}

High ZT

Suppress κ_{ph} 2

G, S and κ_{el} are strongly correlated

$$ZT_{el} = \frac{L_1^2}{L_0L_2 - L_1^2}$$

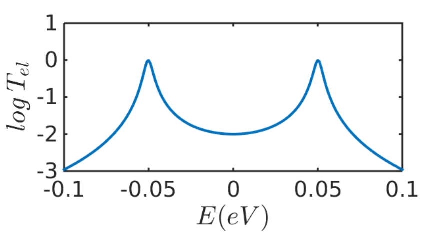
$$L_n = \int_{-\infty}^{+\infty} dE(E - E_F)^n \boxed{T_{el}(E)}(-\frac{\partial f_{FD}}{\partial E})$$

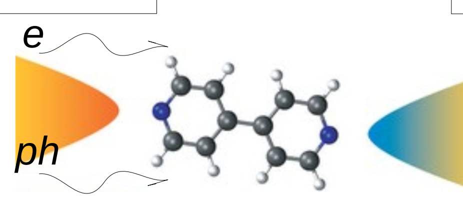
Transmission probability of electrons

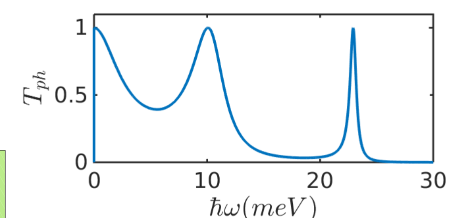
Junction structure dependent

$$\kappa_{ph} = \frac{1}{2\pi}\int_0^{+\infty} d\omega\ \hbar\omega \boxed{T_{ph}(\omega)}(-\frac{\partial f_{BE}}{\partial T})$$

Transmission probability of phonons





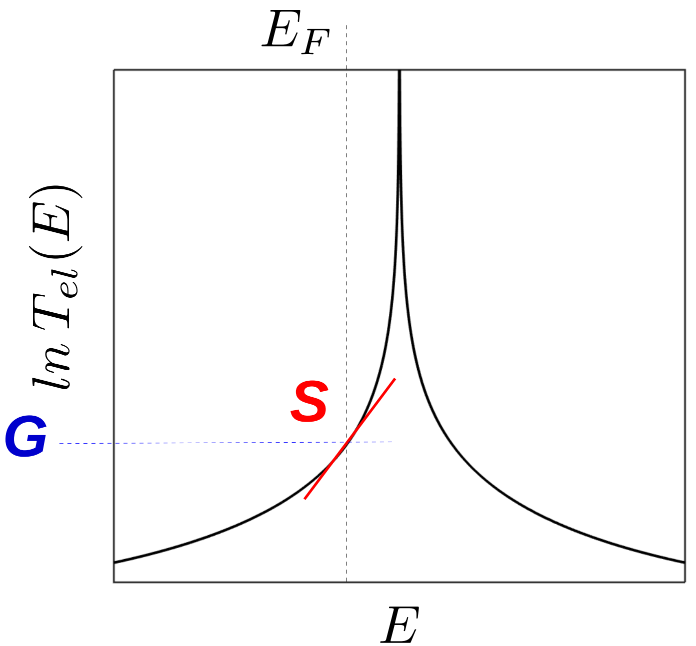


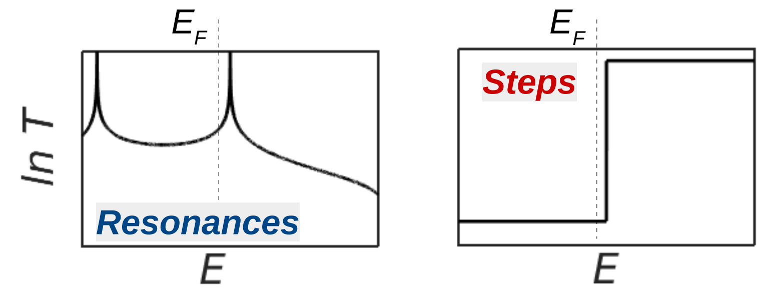
Simultaneous optimisation of T_{el} and T_{ph}

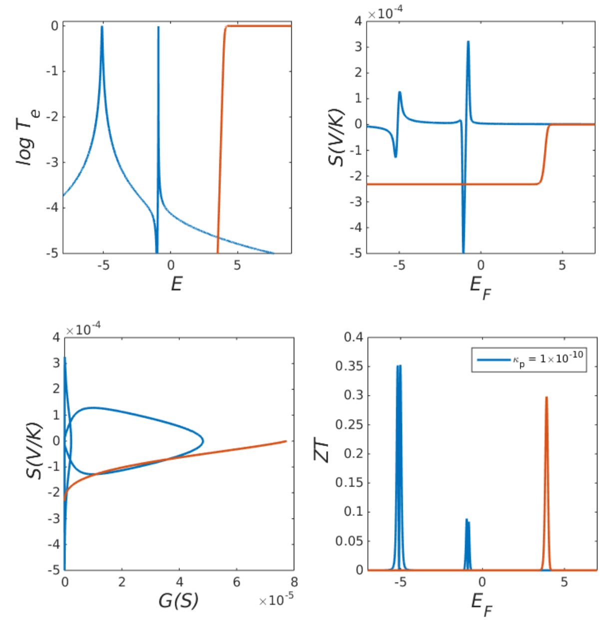
Strategies to maximise ZT_{el}

$$S \approx -\alpha|e|T\frac{d\ln T(E)}{dE}|_{E=E_F}$$

$$G \approx \frac{2e^2}{h}T(E_F) \qquad \kappa_{el} \approx \alpha T G$$

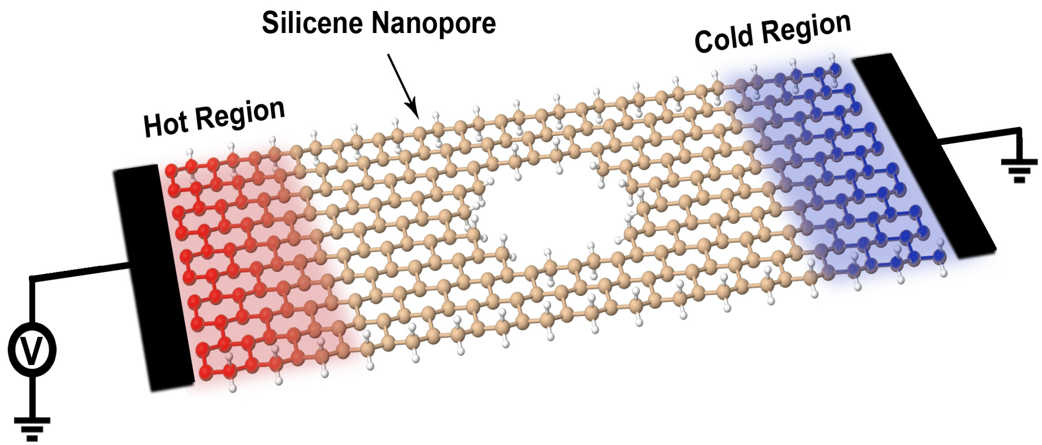


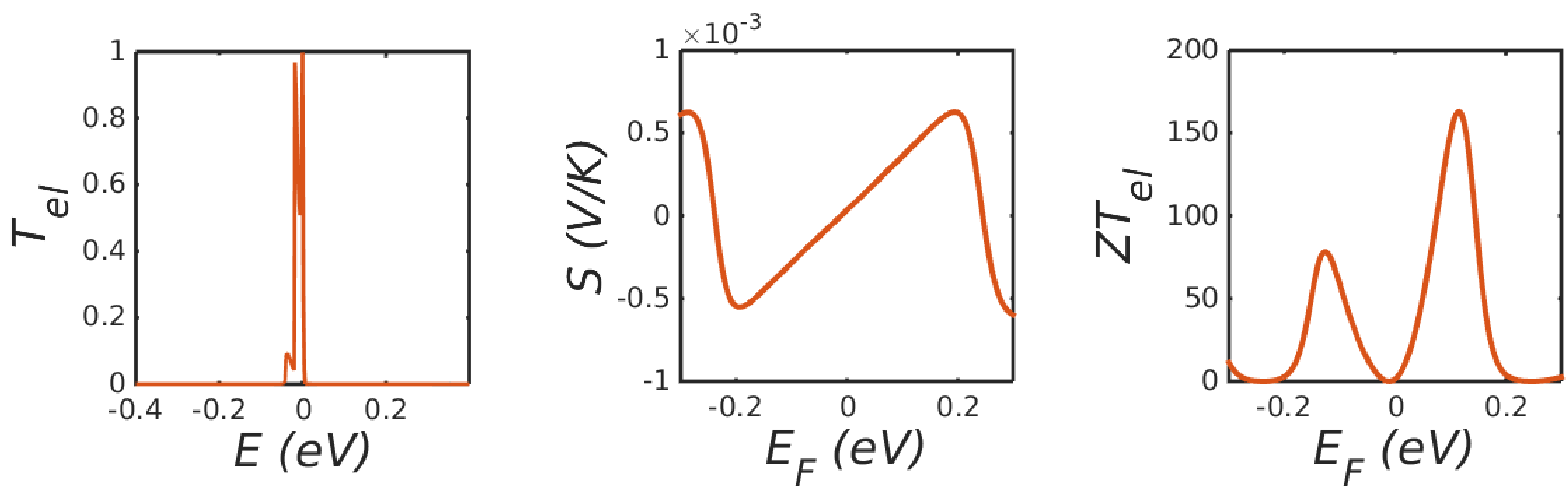




$$ZT_{el} = \frac{GS^2T}{\kappa_{el}} \approx \frac{S^2}{\alpha = 2.44 \times 10^{-8}} \xrightarrow{ZT_{el} > 1} \boxed{S > 160\ \mu V/K}$$

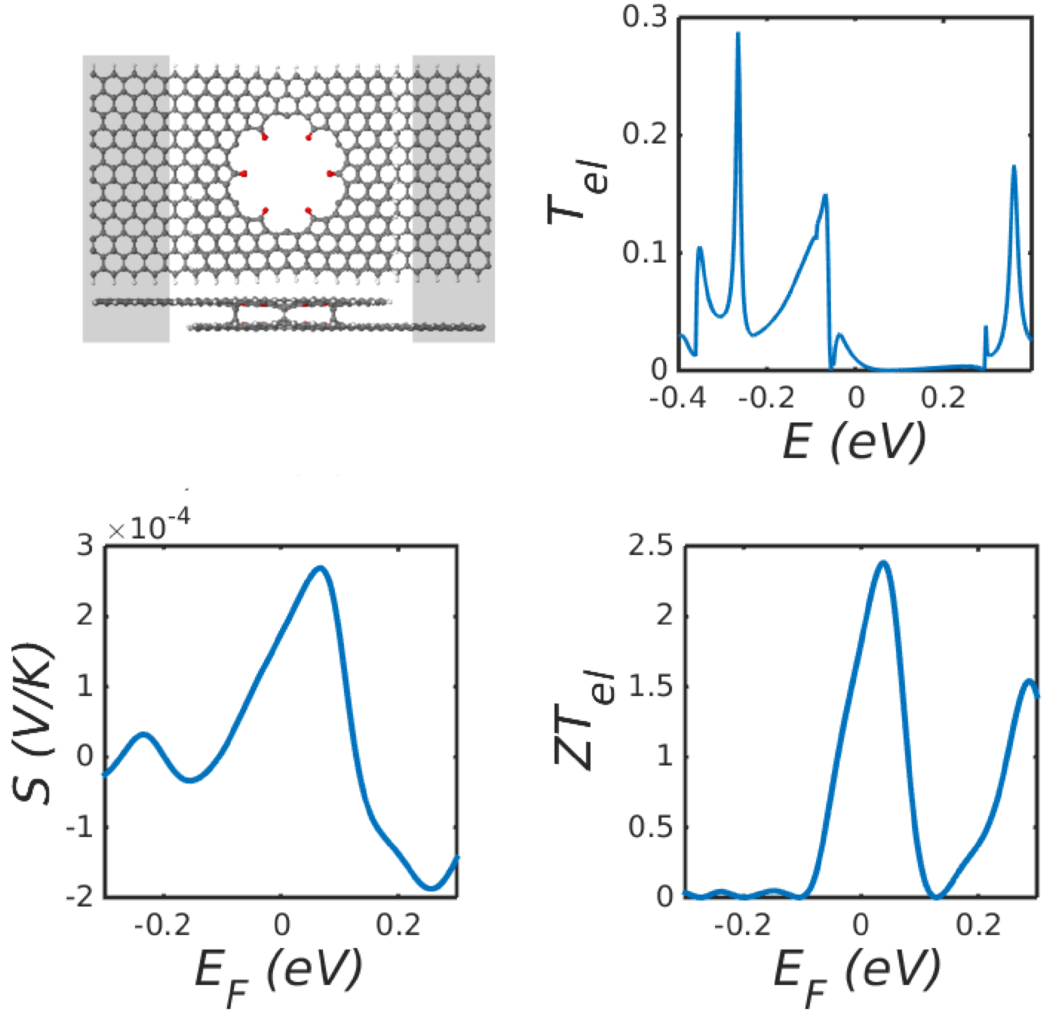
Utilising sharp resonances



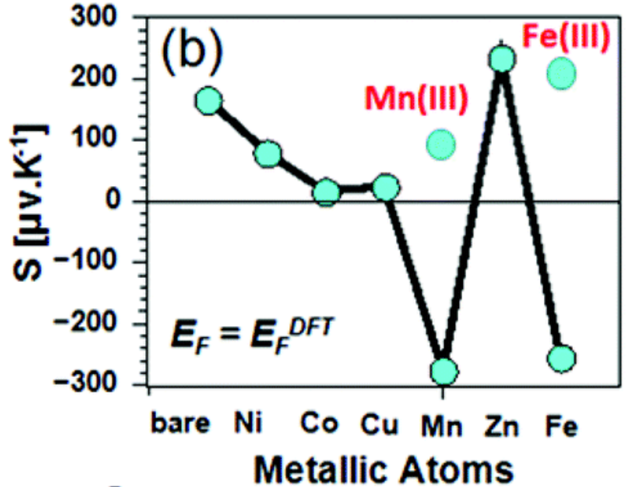


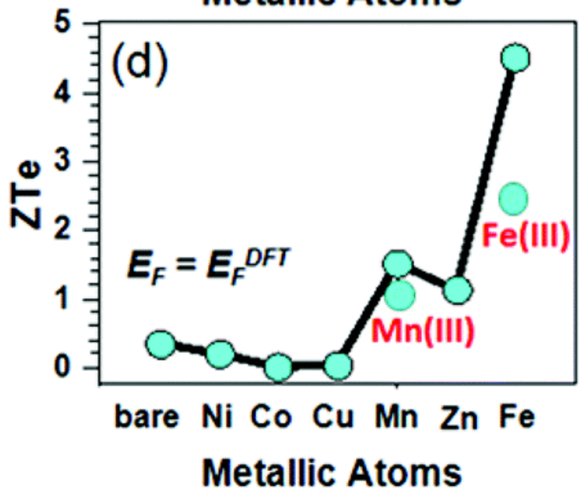
Sadeghi, Sangtarash, Lambert, 2015, Scientific reports 5, 9514

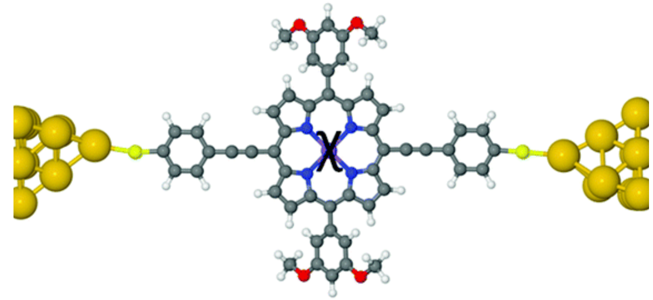
Utilising step functions

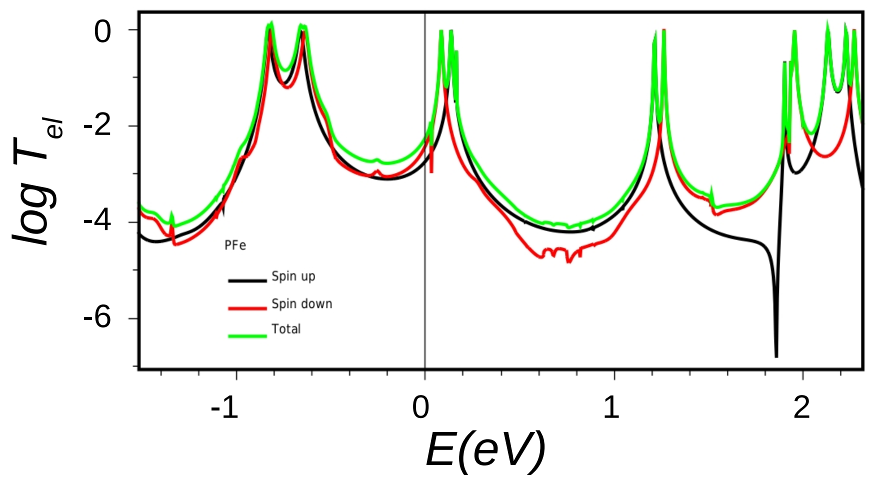


Metallic atom to adjust Fermi energy

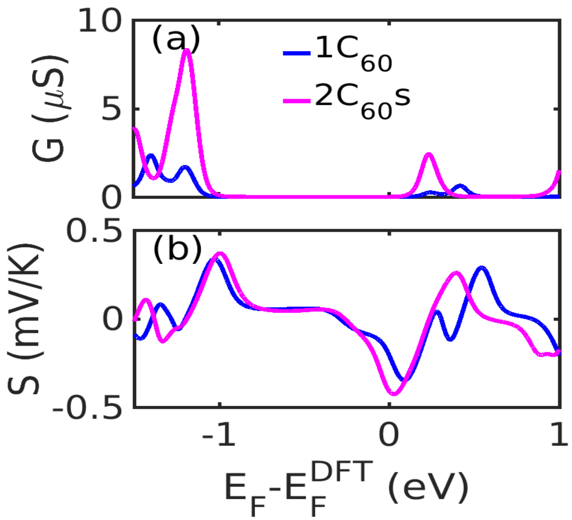


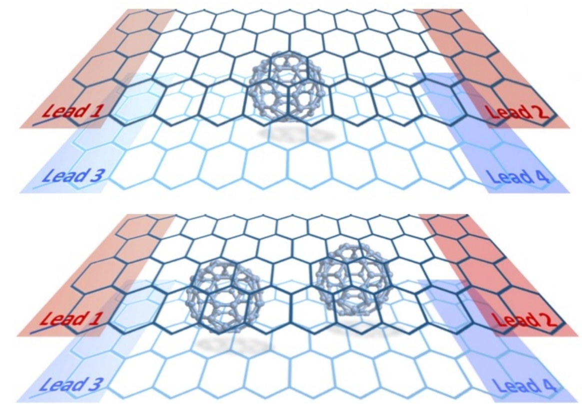




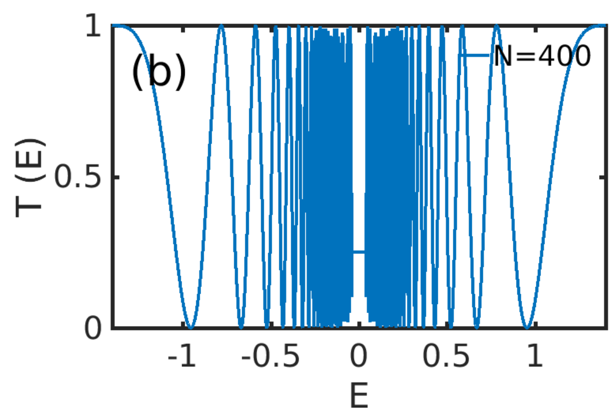


From single molecules to thin films



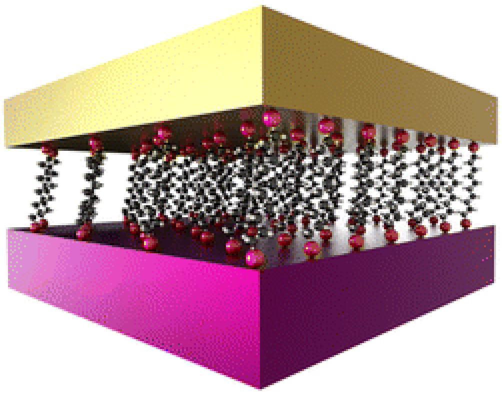


Thermopower does not behave classically

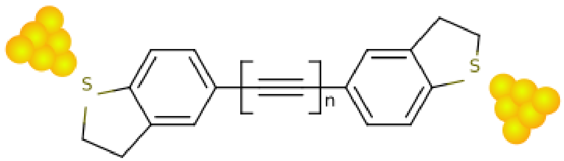


Strategies to suppress κ_{ph}

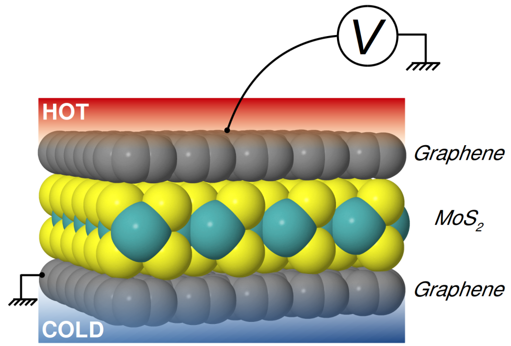
Suppress using mismatch in electrode molecule-interface



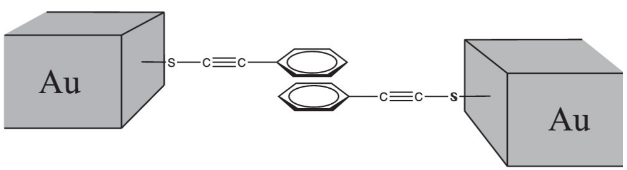
Filter using Low-Debye Frequency electrode



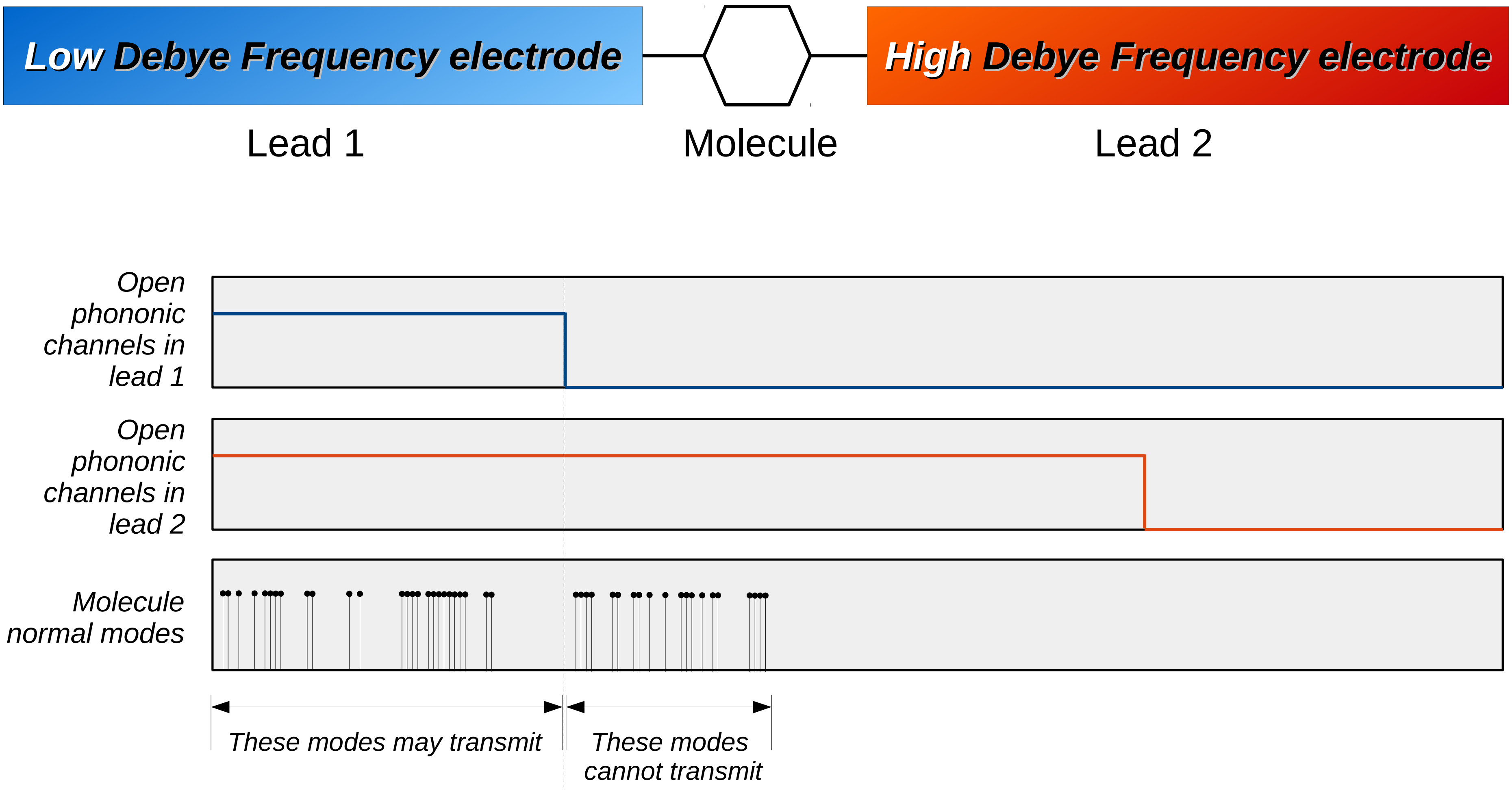
Suppress through interfaces of van der Waals heterostructures



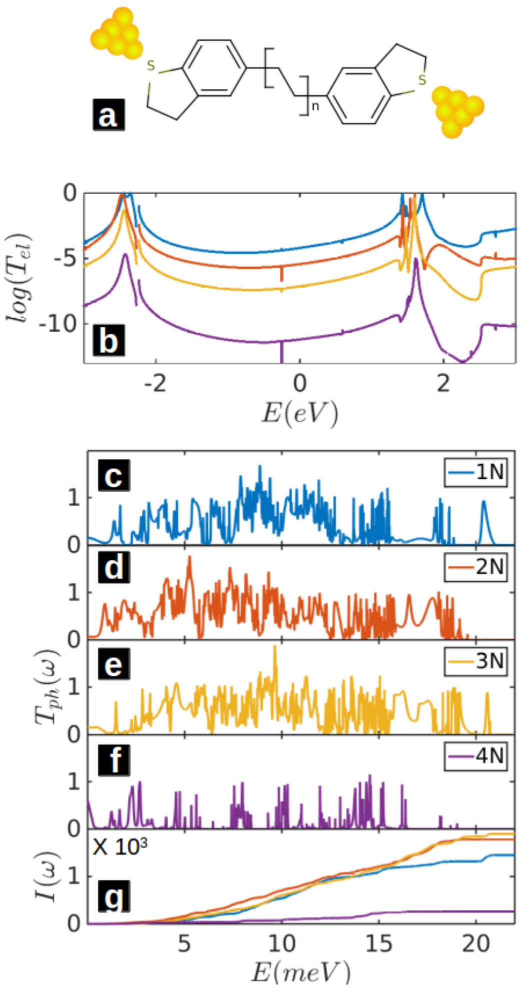
Suppress through π-stacked molecular structure

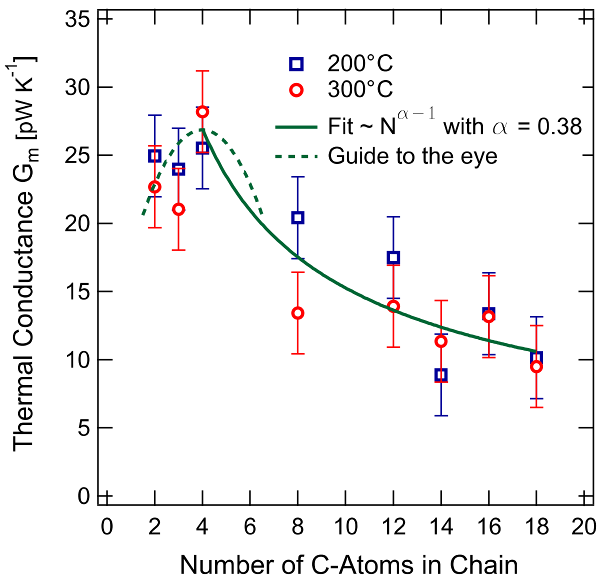


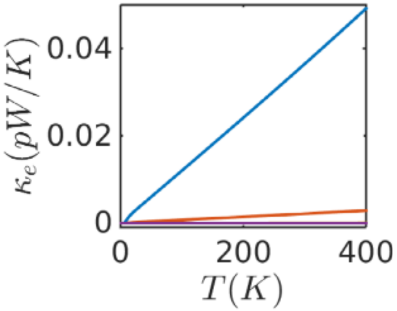
Debye frequency of electrodes

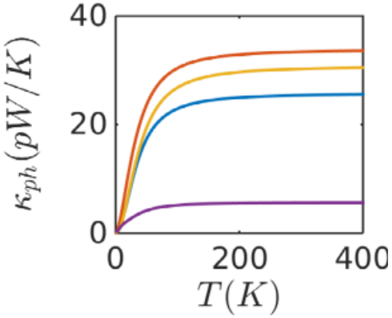


Alkanes with BT anchor & different length

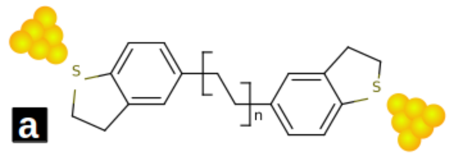


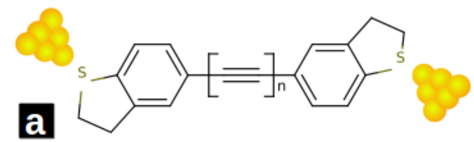


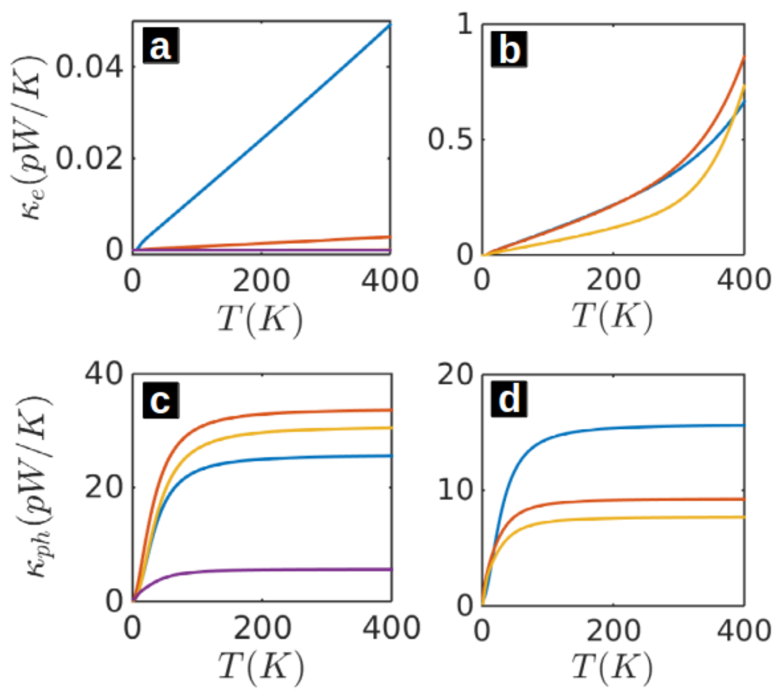


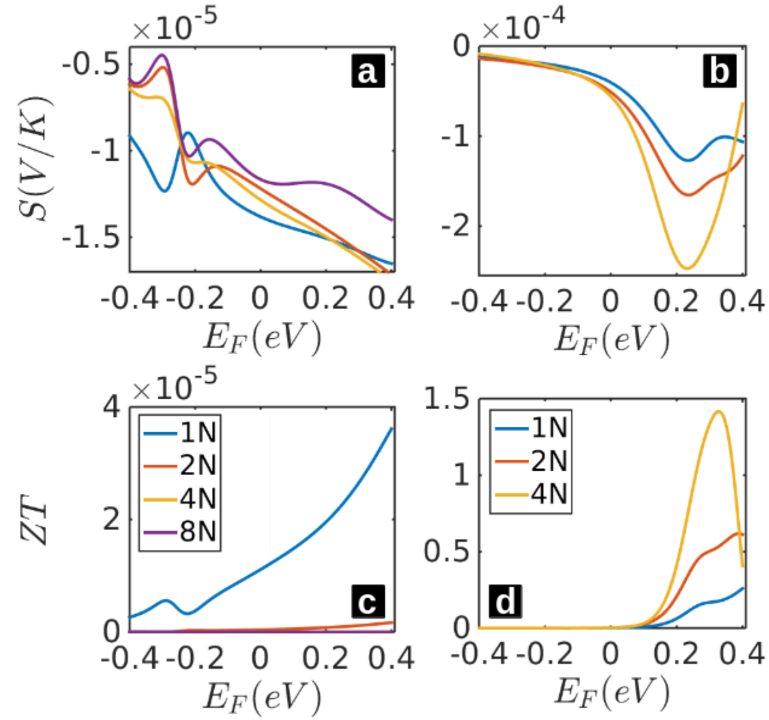


Alkanes vs. oligoynes



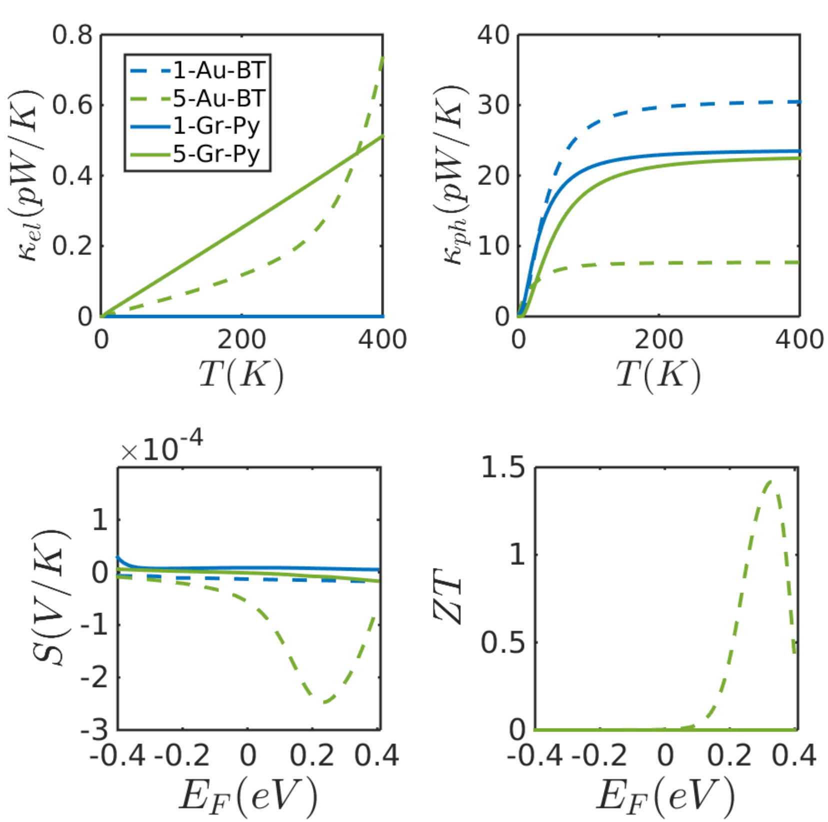


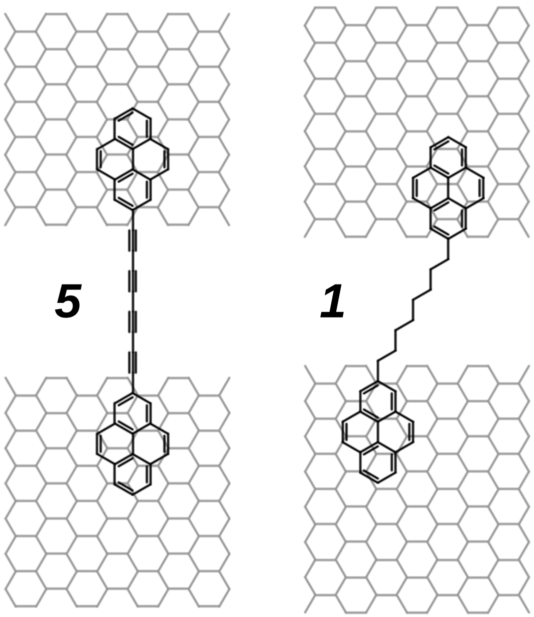




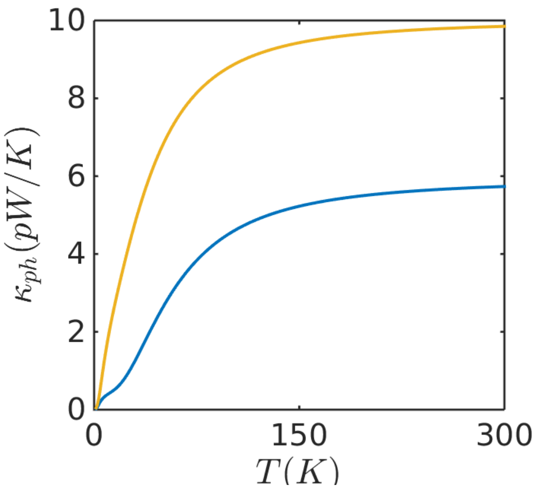
Sadeghi, Sangtarash, Lambert, Nano Lett. 2015, 15, 7467

π-π stacking to suppress phonon transport





Edge-over-edge Zinc-porphyrin



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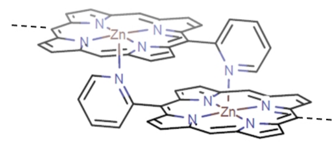

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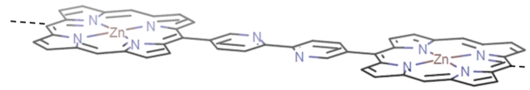
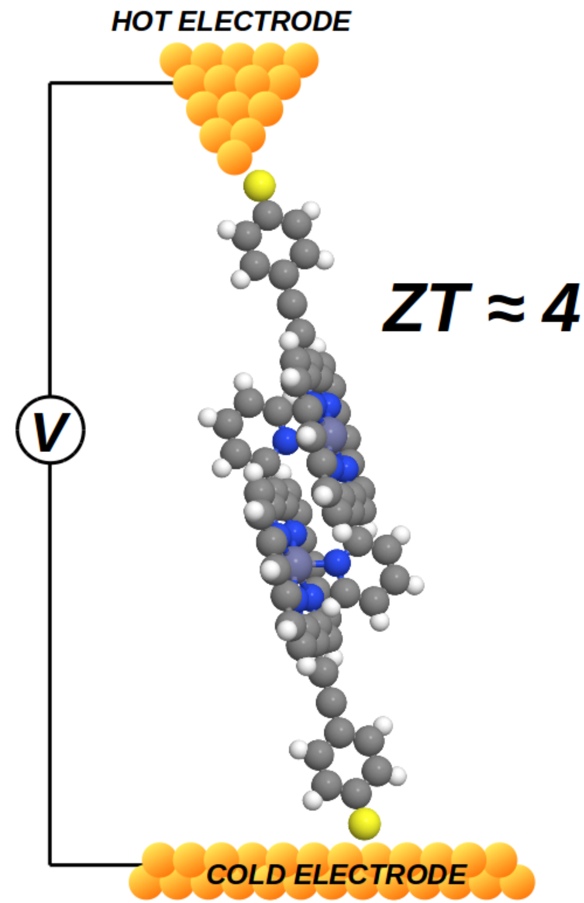
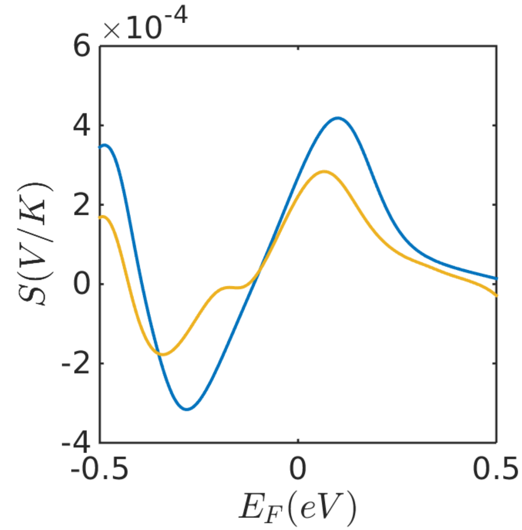
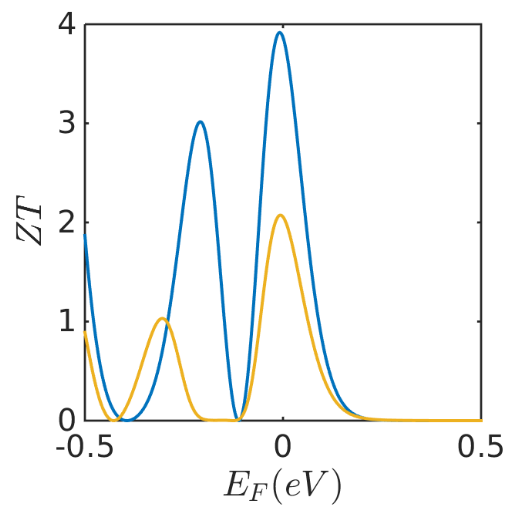
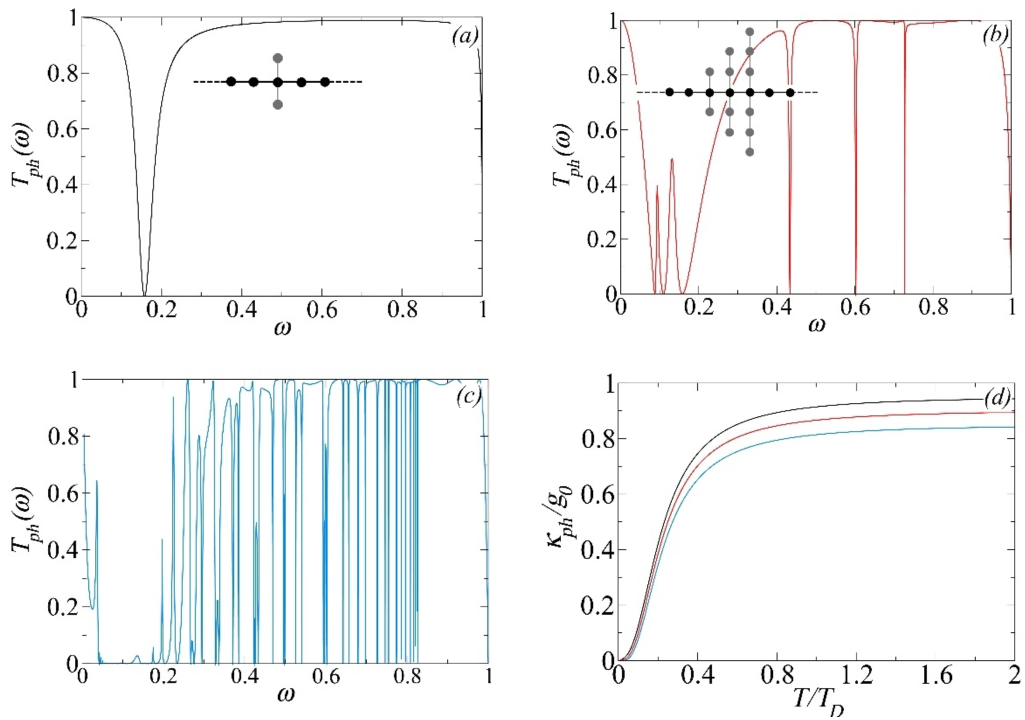


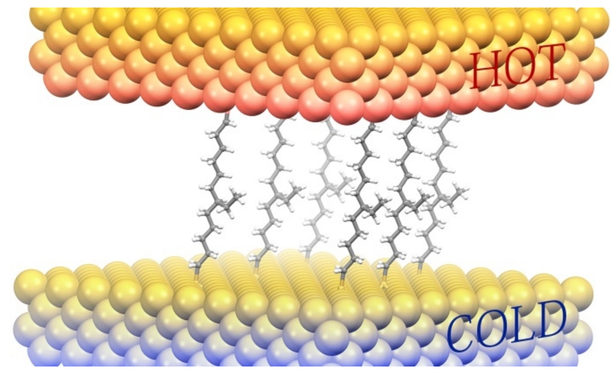






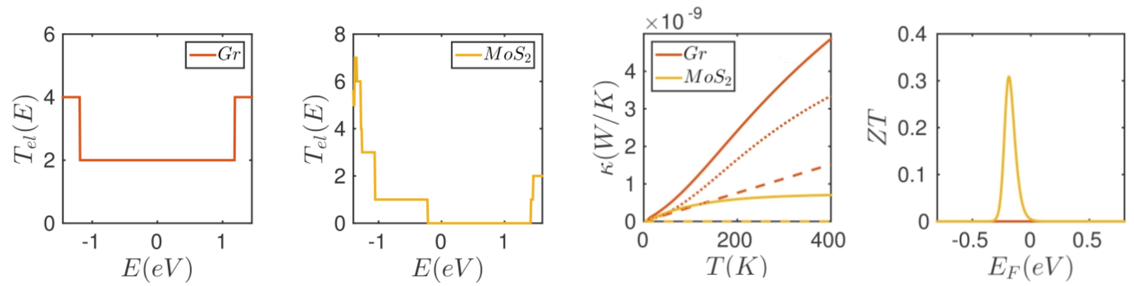
Pendent groups to suppress phonons

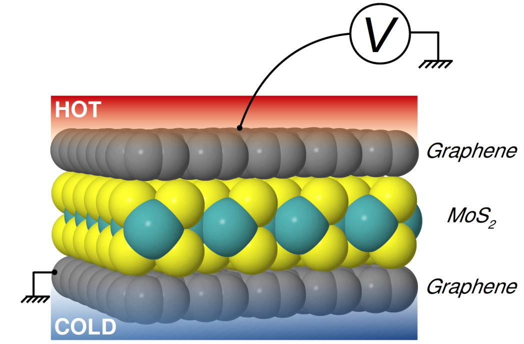




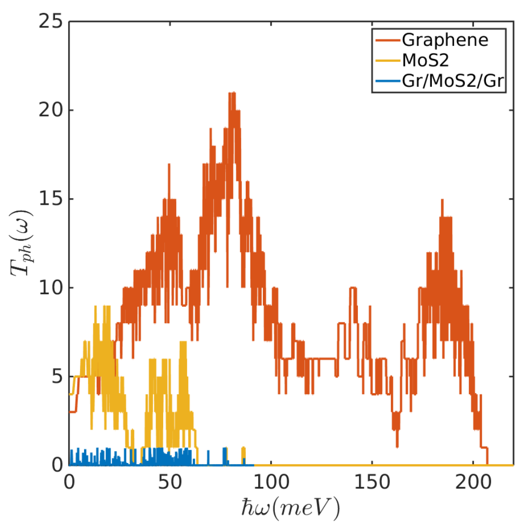
Famili, Grace, Sadeghi, Lambert, 2017

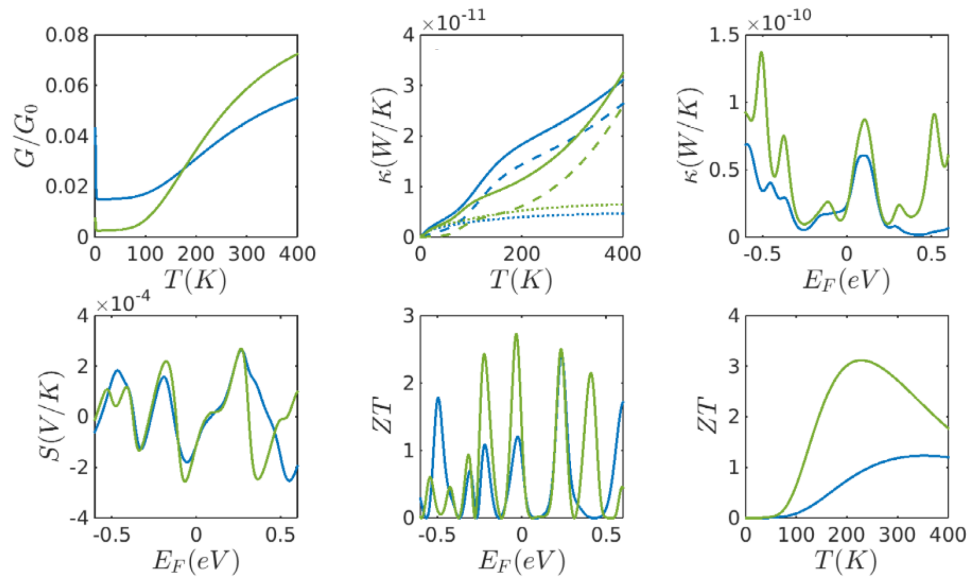
Cross-plane thermoelectricity





vdW heterostructures





Conclusion

- Quantum interference could be utilised to enhance electronic thermoelectric figure of merit ZT_{el}
- QI induced step and Lorentzian like electronic transmission function $T_{el}(E)$ to enhance ZT_{el}
- Phonon interference could be utilised to suppress phononic thermal conductance.
- Debye frequency of electrodes, molecule-electrode interface, pendent groups, molecular velcros, π-π stacking and cross plane transport in vdW heterostructures could be used to suppress phonons.
- These could be combined to some extend to obtain high ZT.

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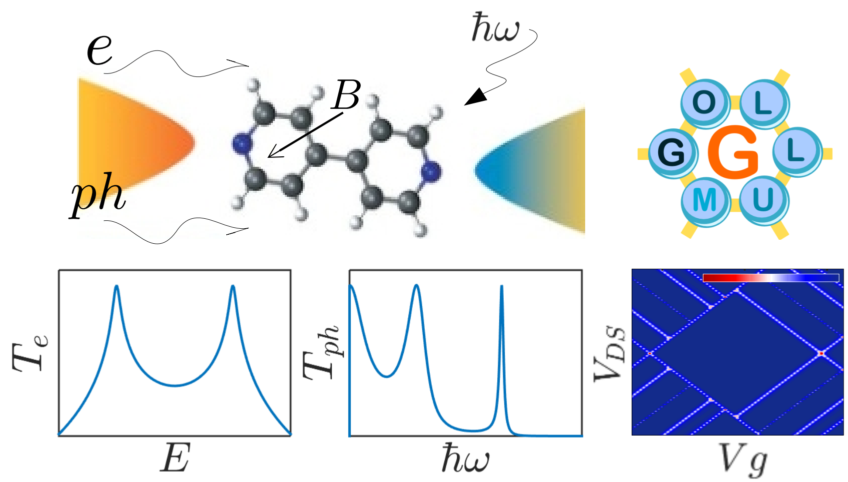
Oviedo University, Spain



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Thank you for your attention

Q & A

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