Enhanced thermal transport in multilayered devices arising from electronic charge reconstruction

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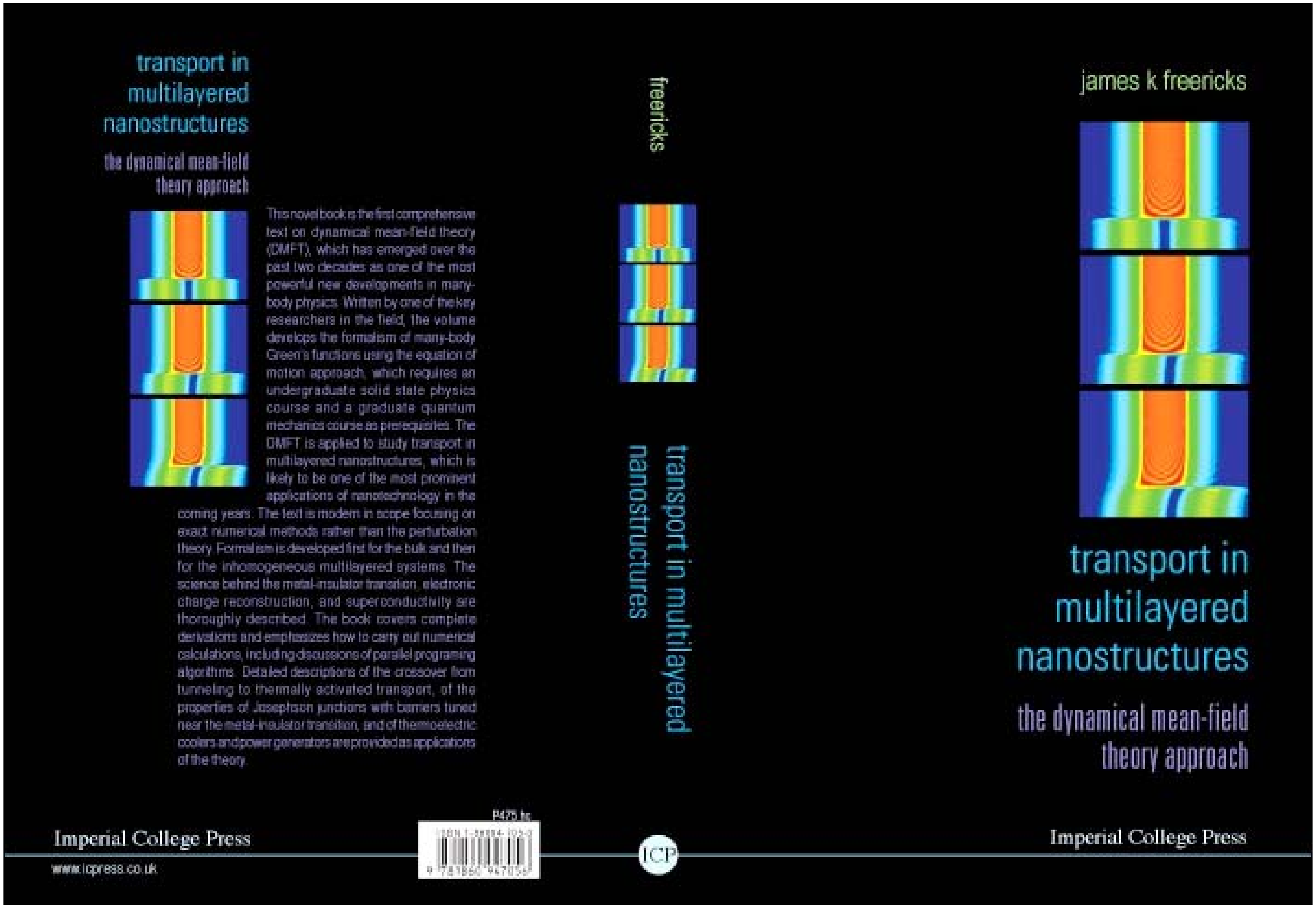
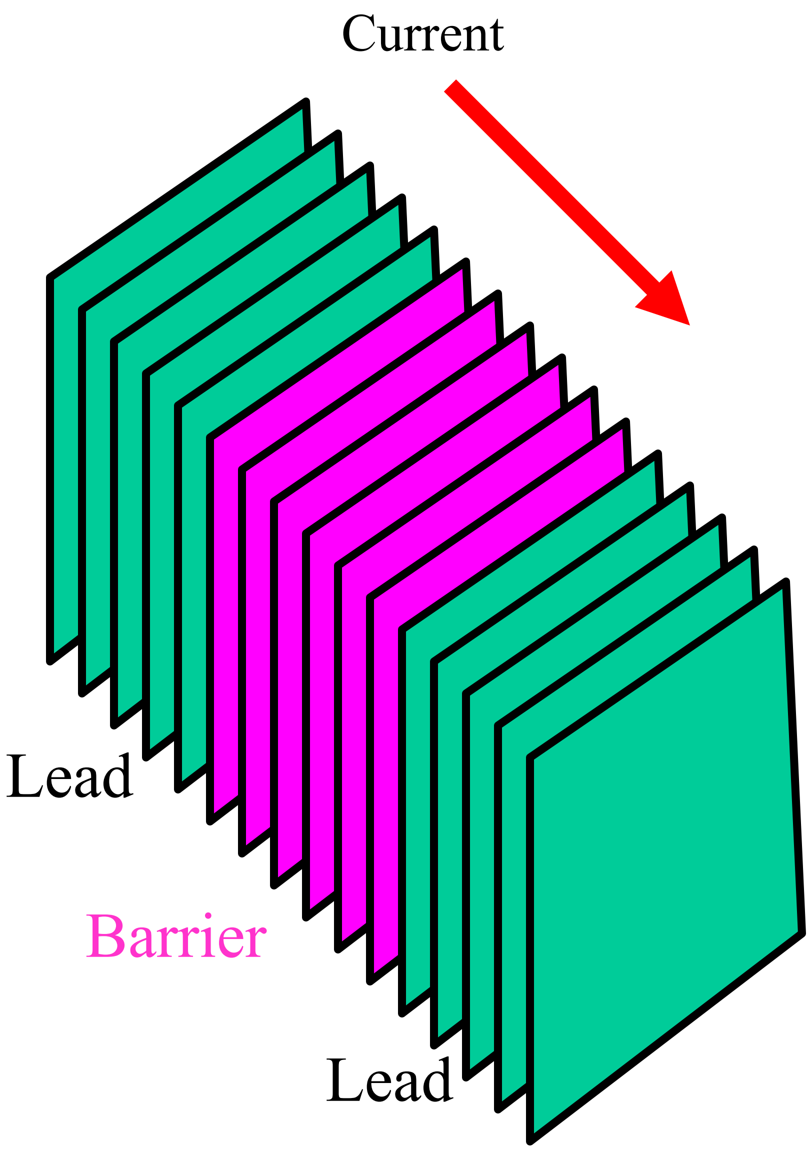


Our model



Device is made from stacking multilayers of different materials and passing current perpendicular to the planes.

The metallic leads can be ballistic or diffusive metals, strongly correlated metals, Kondo systems, heavy Fermions, etc.

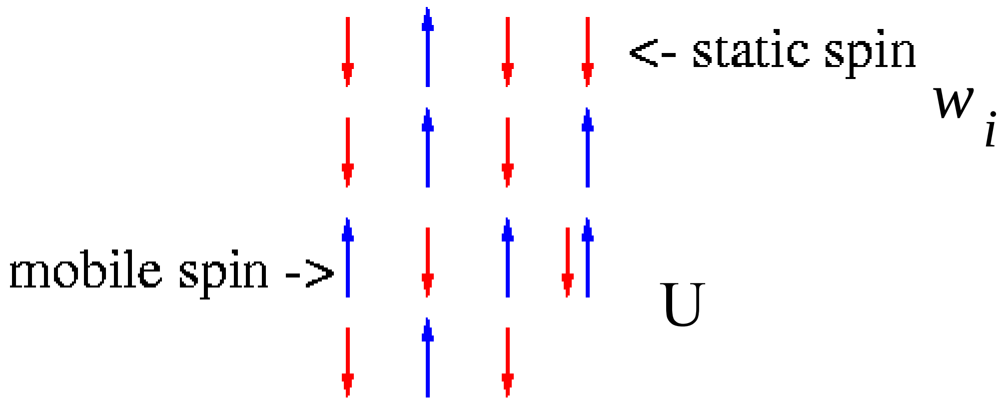
Scattering in the barrier is included via charge scattering with “defects” (as in the Falicov-Kimball model)

Need a theory that can incorporate all forms of transport (ballistic, diffusive, incoherent, and strongly correlated) on an equal footing

A self-consistent recursive Green's function approach called **inhomogeneous dynamical mean field theory** (developed by Potthoff and Nolting) can treat all of these different kinds of transport.

Spinless Falicov-Kimball Model

$$H = -\frac{t}{2\sqrt{d}} \sum_{\langle i,j \rangle} c_i^{\dagger} c_j + E \sum_i w_i + U \sum_i c_i^{\dagger} c_i w_i$$

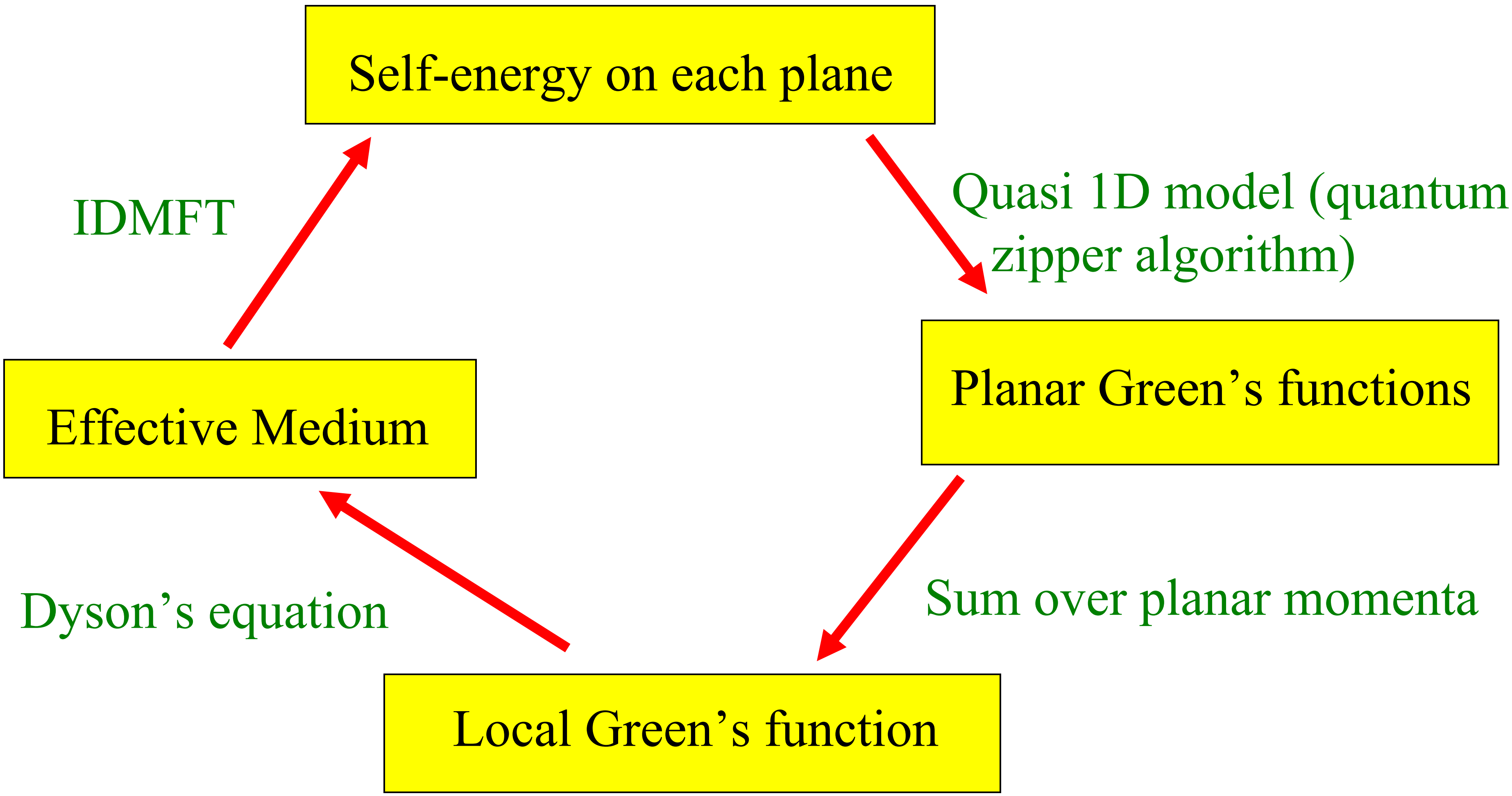


Exactly solvable model in the local approximation using dynamical mean field theory.

Possesses homogeneous, commensurate/incommensurate CDW phases, phase segregation, and **metal-insulator transitions**.

Does not have Fermi-liquid phases, but can describe many strong correlation properties that do not rely on spin-dependent effects.

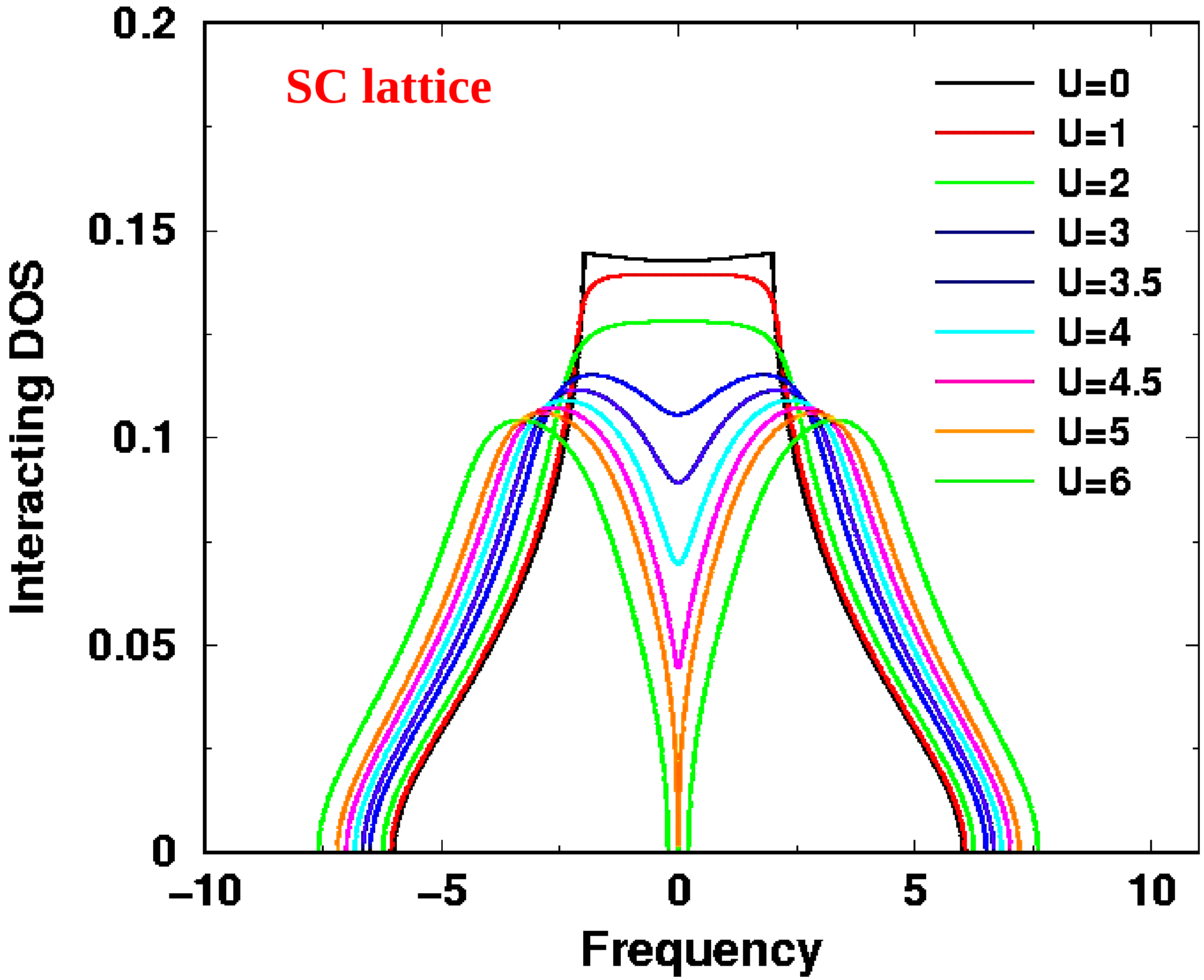
Computational Algorithm



Algorithm is iterated until a self-consistent solution is achieved

Half-filling and the particle-hole symmetric metal-insulator transition …

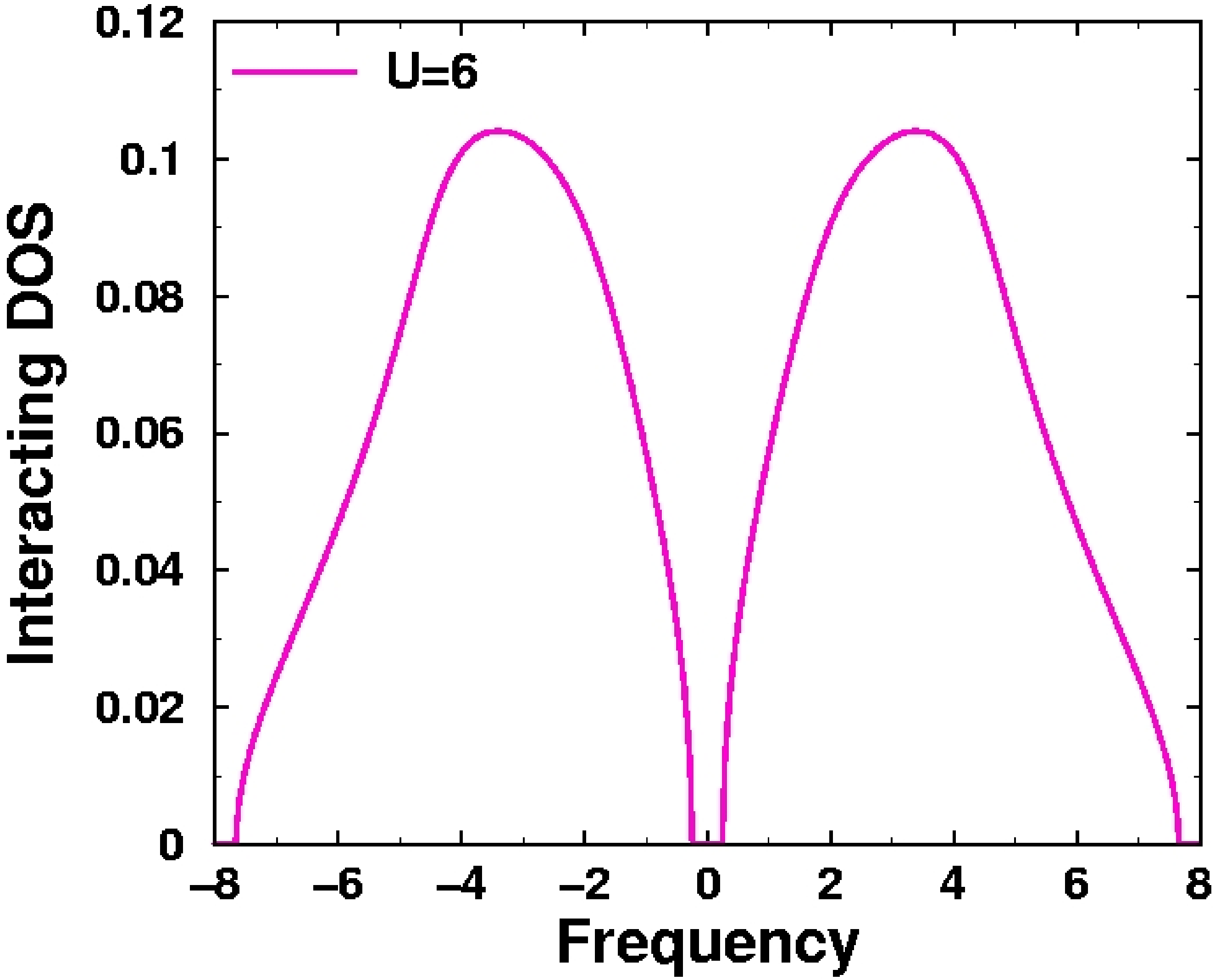
Metal-insulator transition (half-filling)



The Falicov-Kimball model has a **metal-insulator transition** that occurs as the correlation energy U is increased. The bulk interacting DOS shows that a **pseudogap** phase first develops followed by the opening of a **true gap** above U=4.9 (in the bulk).

Note: the FK model is **not a Fermi liquid** in its metallic state since the lifetime of excitations is finite.

Near the MIT (U=6)



If we take t=0.25ev, then W=3ev, and the gap size is about 100mev.

This is a correlated insulator with a small gap, close to the MIT.

Charge transport

Junction resistance

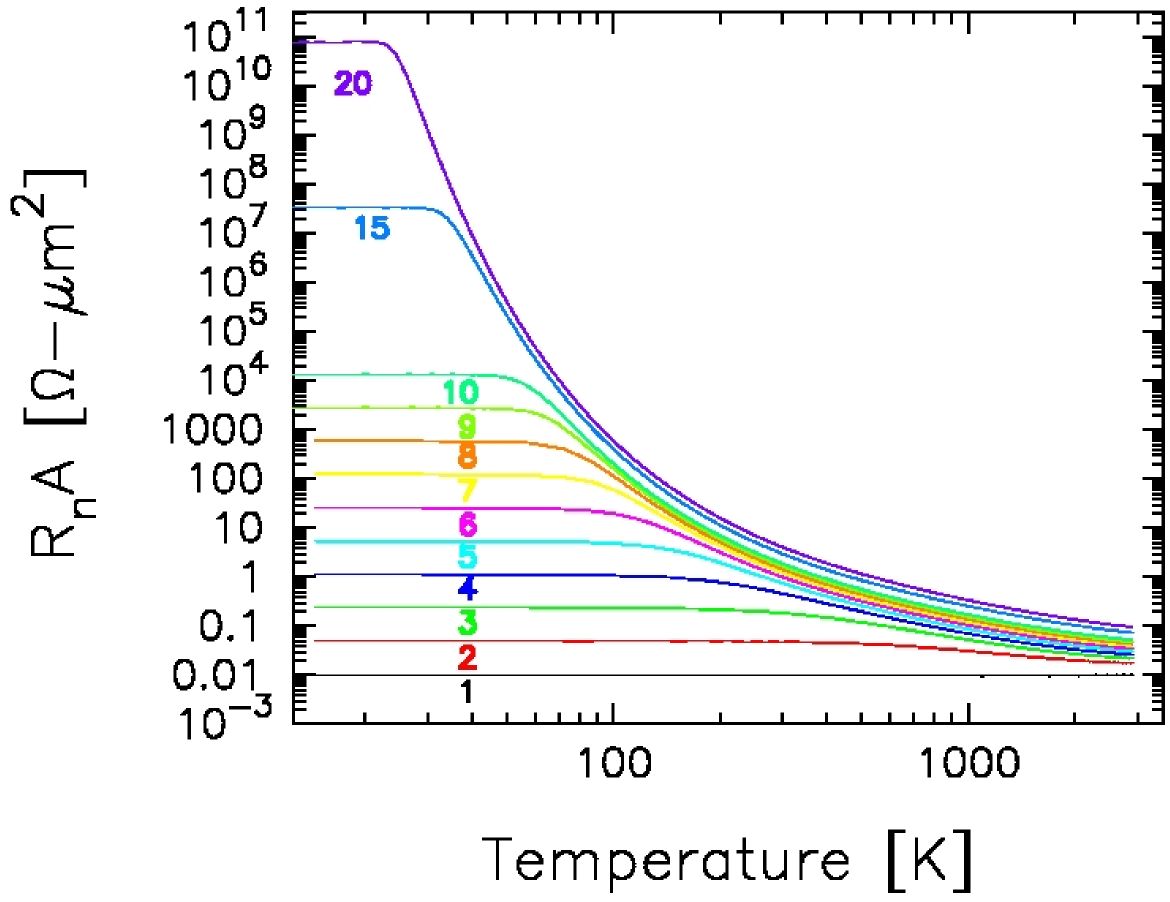
- The linear-response resistance can be calculated in equilibrium using a Kubo-Greenwood approach.
- We must work in real space because there is no translational symmetry.
- R_n is calculated by inverting the isothermal conductivity matrix and summing all matrix elements of the inverse.

Junction resistance (derivation)

- Maxwell's equation gives $\mathbf{j}_i=\sum_j \sigma_{ij}\mathbf{E}_j$ where the index denotes a plane in the layered device. (The field at plane j causes a current at plane i.)
- Taking the matrix inverse gives $\mathbf{E}_i=\sum_j \sigma^{-1}_{ij}\mathbf{j}_j$; but the current is conserved, so **j** does not depend on the planar index.
- Calculating the voltage gives $V=a\sum_i E_i=a\sum_{ij}\sigma^{-1}_{ij}j$, so the resistance-area product is $R_nA=a\sum_{ij}\sigma^{-1}_{ij}$

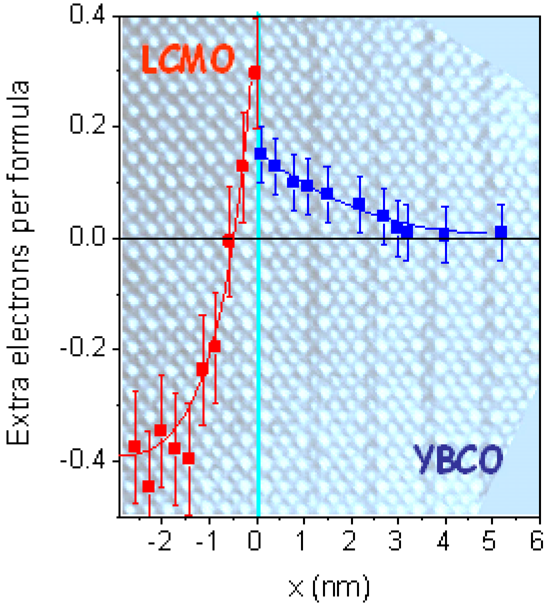
Resistance for U=6 (correlated insulator)

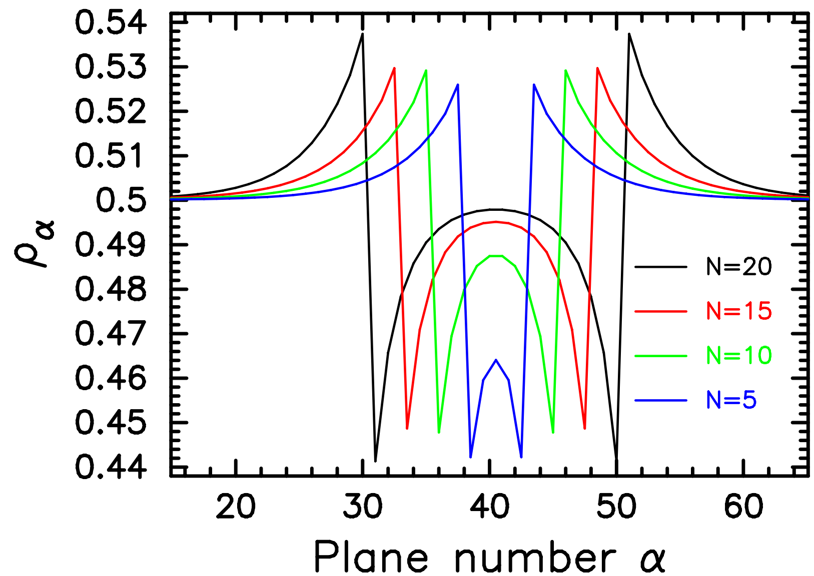
Resistance here shows the tunneling plateaus clearly, and a strong temperature dependence in the incoherent regime.



Particle-hole asymmetry is necessary for thermoelectric devices …

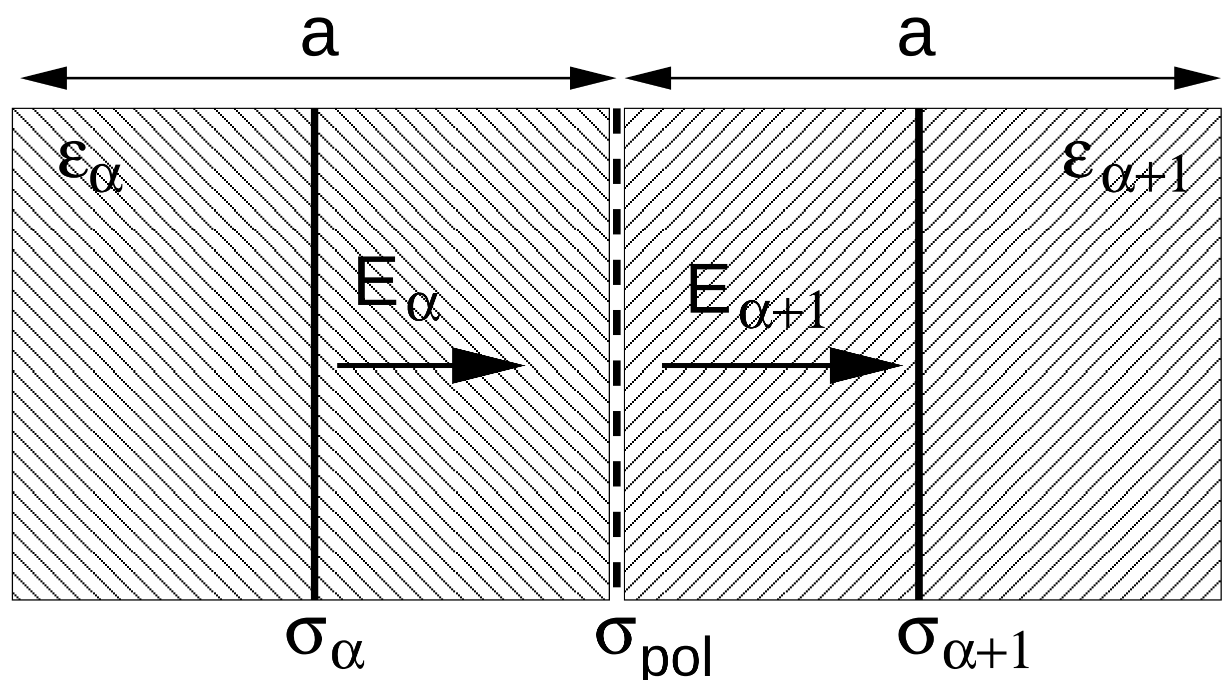
Electronic charge reconstruction





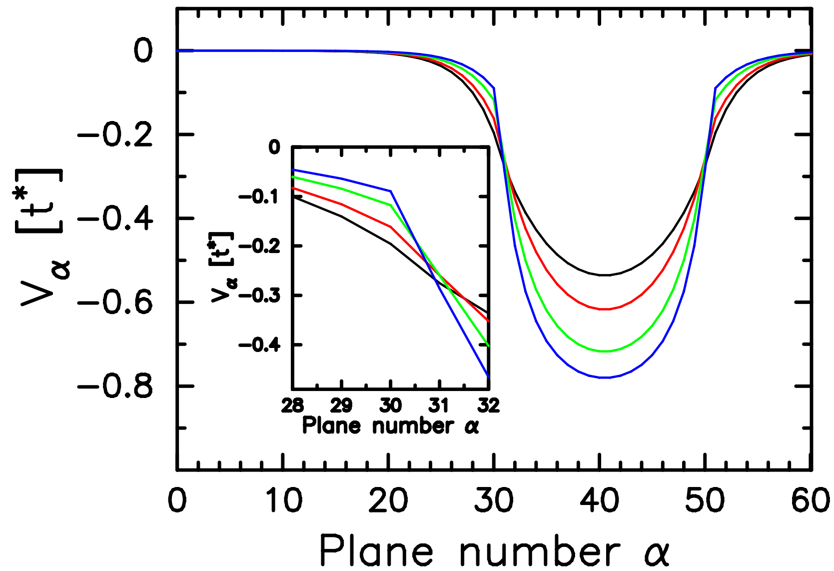
Using a scanning transmission electron microscope with electron energy-loss spectroscopy, one can directly measure the electronic charge at each plane of a strongly correlated multilayered nanostructure. Left are experimental results by *Varela et al.* on YBCO/LCMO heterostructures, right is a simple theory for strongly correlated nanostructures.

Theoretical treatment

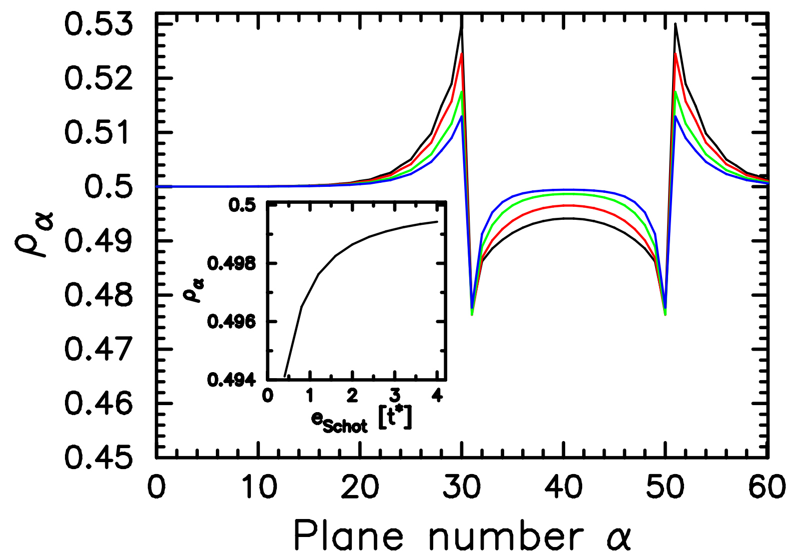


We employ a semiclassical treatment to handle the electronic charge reconstruction. We allow charge to be rearranged on different planes, as determined by the electrochemical potential at a given plane site, and then determine the classical Coulomb potential from planes of net charge, with dielectric constants that can vary from plane to plane.

Coulomb potential

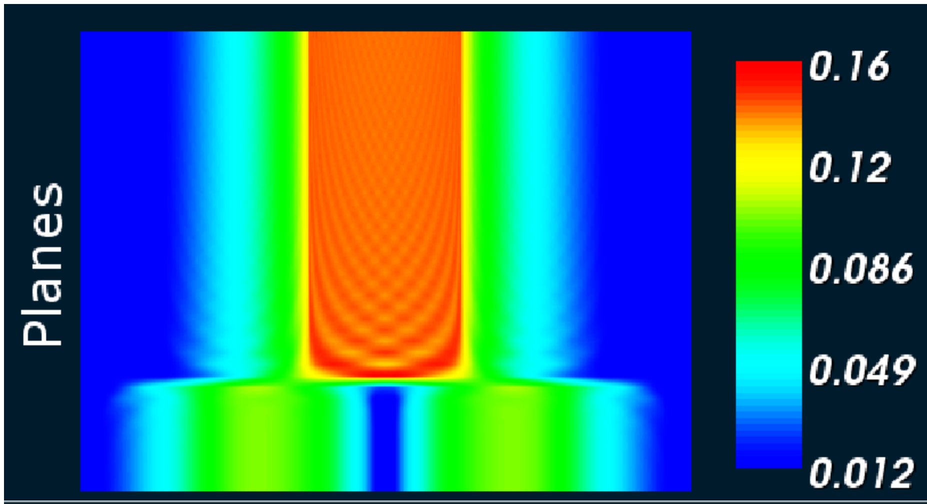


The Coulomb potential develops a kink at locations where the dielectric constant changes (i.e. at the interfaces), and it goes to zero far from the interface due to overall conservation of charge.

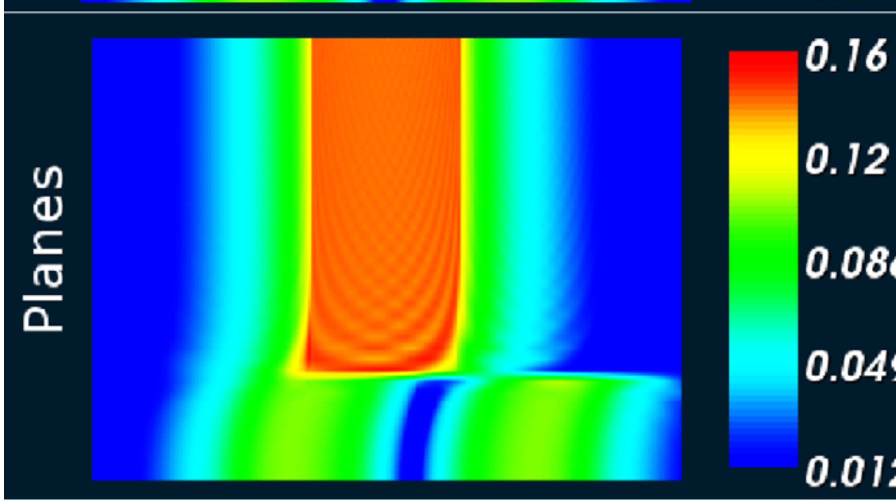


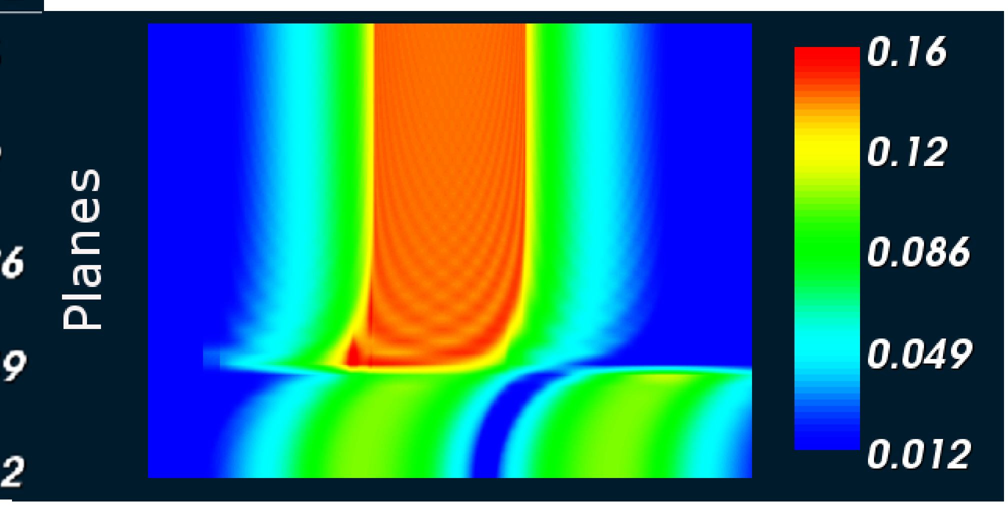
As the screening length decreases, the total charge that is rearranged gets smaller for a fixed chemical potential mismatch of the bulk materials.

DOS with electronic charge reconstruction



Changing the band offsets creates particle-hole asymmetry in the DOS.





Thermal transport in a multilayered nanostructure

Heat Current Conservation

- Unlike the charge current, the heat current need not be conserved in a multilayered nanostructure.
- The experimental conditions will determine the boundary conditions for the heat current, which need to be employed to solve for the heat transport.
- We describe the Seebeck effect here.

Heat transport equations

In the presence of field and temperature gradients, the charge and heat currents satisfy:

$$j_i = e^2 \sum_j L^{11}_{ij} E_j - e \sum_j L^{12}_{ij} (T_{j+1} - T_{j-1})/2a$$

$$j_{Qi} = \sum_j L^{21}_{ij} E_j - \sum_j L^{22}_{ij} (T_{j+1} - T_{j-1})/2a$$

Where the L matrices are found from the **Jonson-Mahan theorem** (current and heat-current correlation functions in real space)

Seebeck effect

In the Seebeck effect, we isolate the device and work with an open circuit. *Hence there is no heat created or destroyed in the steady state (i.e., the heat current is conserved) and the total charge current vanishes*:

The E field becomes $E_j = \sum_{jk} (L^{11})^{-1}{}_{ij} L^{12}{}_{jk} (T_{k+1} - T_{k-1})/2a$

The temperature gradients become

$\sum_j M^{-1}{}_{ij} j_Q = -(T_{i+1} - T_{i-1})/2a$; $M = -L^{21}(L^{11})^{-1}L^{12} + L^{22}$

Hence, $\Delta T = -\sum_{ij} M^{-1}{}_{ij} j_Q$, $\Delta V = -a\sum_{ij}[(L^{11})^{-1}L^{12}M^{-1}]_{ij} j_Q$, and the Seebeck coefficient is

$$S = \Delta V/\Delta T = a\sum_{ij}[(L^{11})^{-1}L^{12}M^{-1}]_{ij} / \sum_{ij} M^{-1}{}_{ij} \text{ [in units of } |e|/k_B]$$

Note the weighting by the matrix M, which is different for a nanostructure than in the bulk, where that factor cancels as can be seen from the convolution theorem!

Thermal transport created from electronic charge reconstruction

Seebeck effect

Numerically we evaluate the Seebeck coefficient for two particle-hole symmetric bulk materials with an electronic charge reconstruction. **The Seebeck effect can become quite large!**

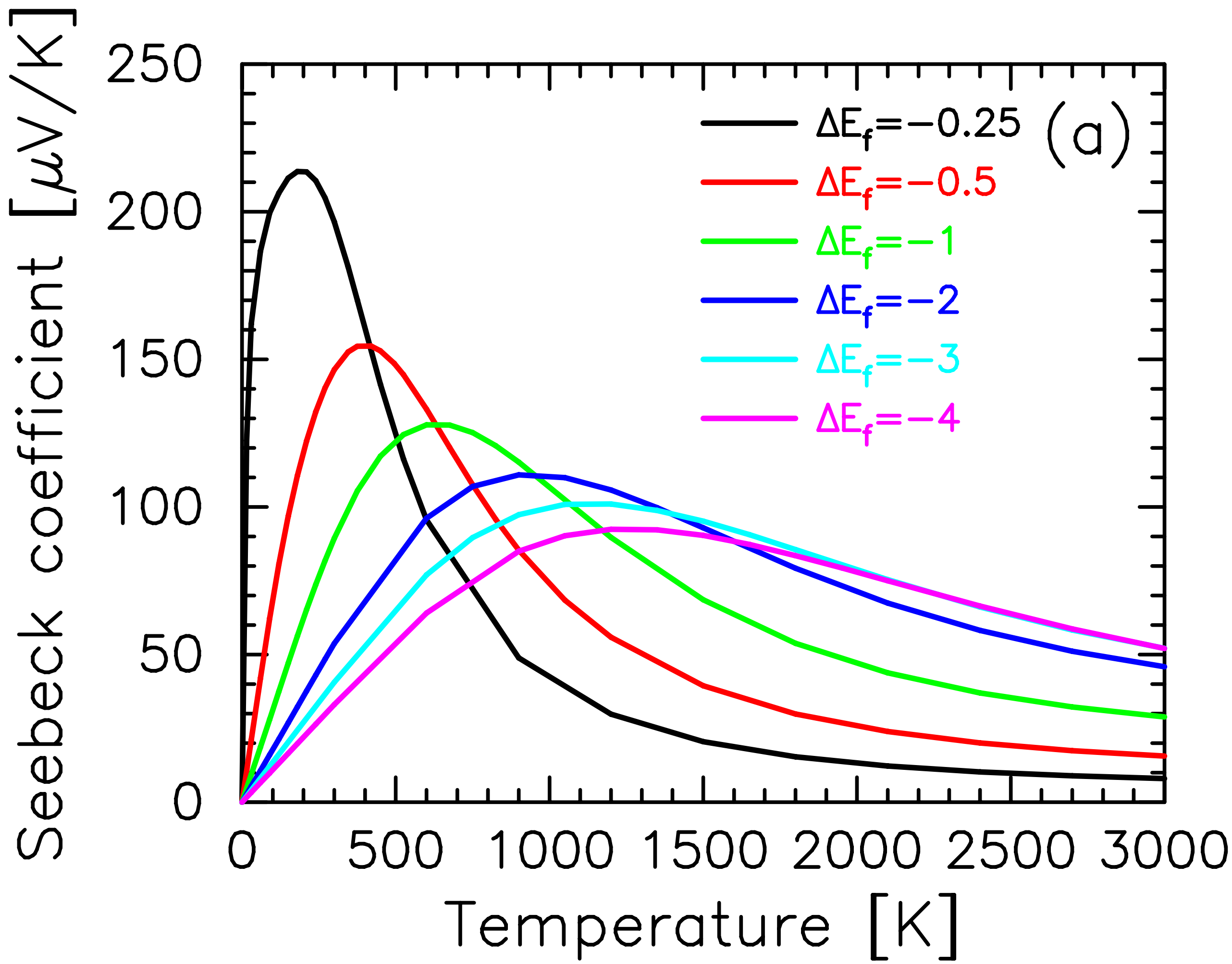
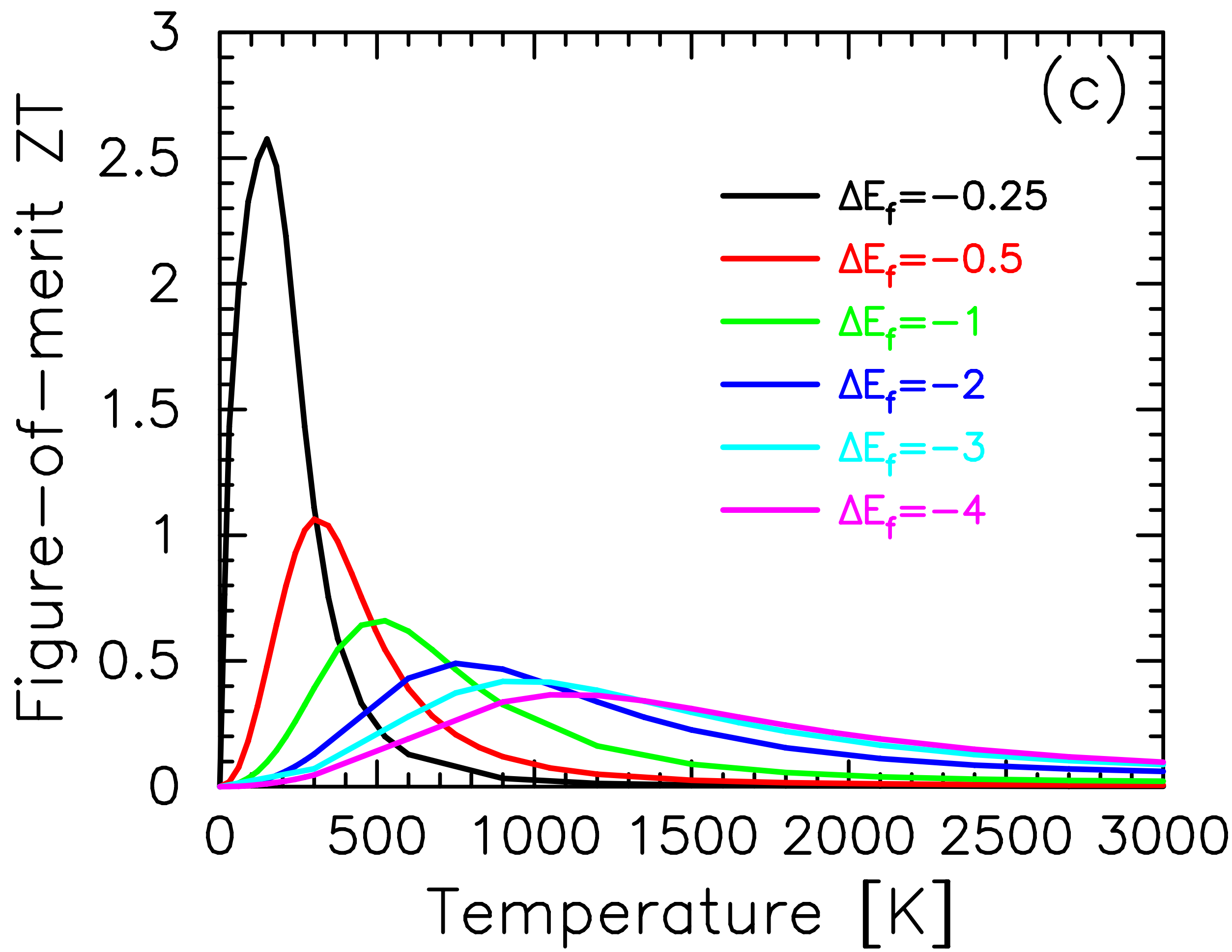


Figure of merit

The figure-of-merit can also become large, and **is bigger than 1 for small band offsets**. *The phonon thermal conductance can dramatically reduce the figure-of-merit though.*



Conclusions

Using multilayers of different materials creates a new means for tuning electronic transport and thereby optimizing transport properties, especially for thermoelectric properties.

The parameter space is huge, because it may not be optimal to use good bulk materials in the multilayered device, rather electronic charge reconstruction can help produce new kinds of behavior.

This is a realm where efficient computation is valuable in trying to narrow down the parameter space, determine design rules for optimization of performance, and find real material candidates for fabrication and testing.