Transport in strongly correlated multilayered nanostructures: *the dynamical mean-field theory approach*

J. K. Freericks

Department of Physics, Georgetown University, Washington, DC 20057

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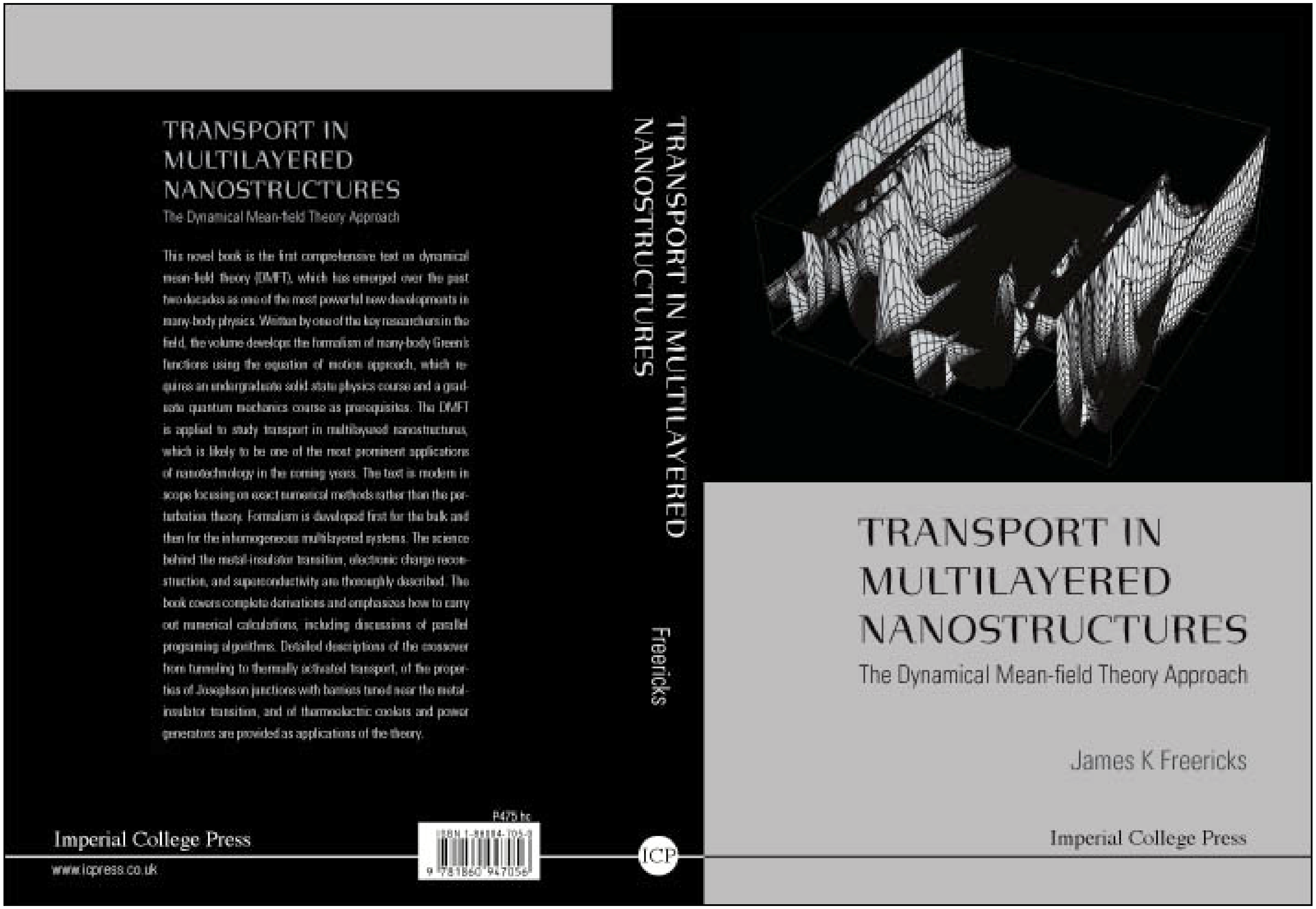
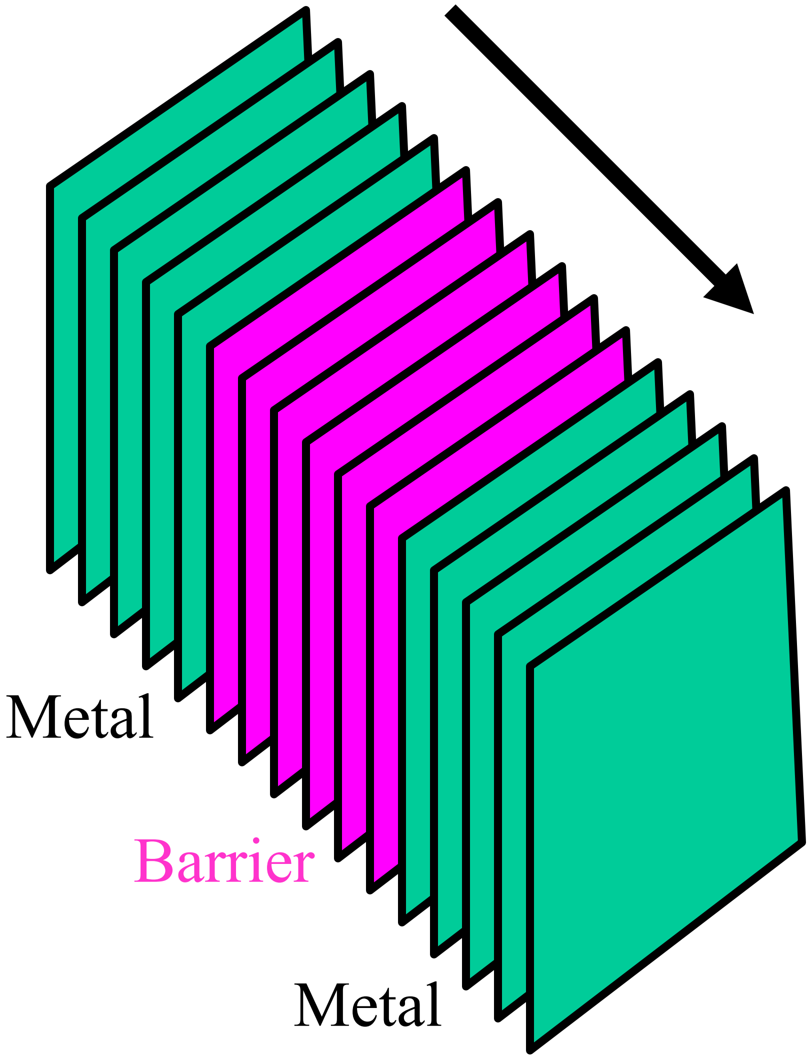


Multilayered nanostructures as devices



- Sandwich of metal-barrier-metal with current moving perpendicular to the planes
- Nonlinear current-voltage characteristics
- Josephson junctions, diodes, thermoelectric coolers, spintronic devices, etc.
- Band insulators: AlO_x MgO
- Correlated materials: FeSi, $SrTiO_3$
- Near MIT: V_2O_3, Ta_xN

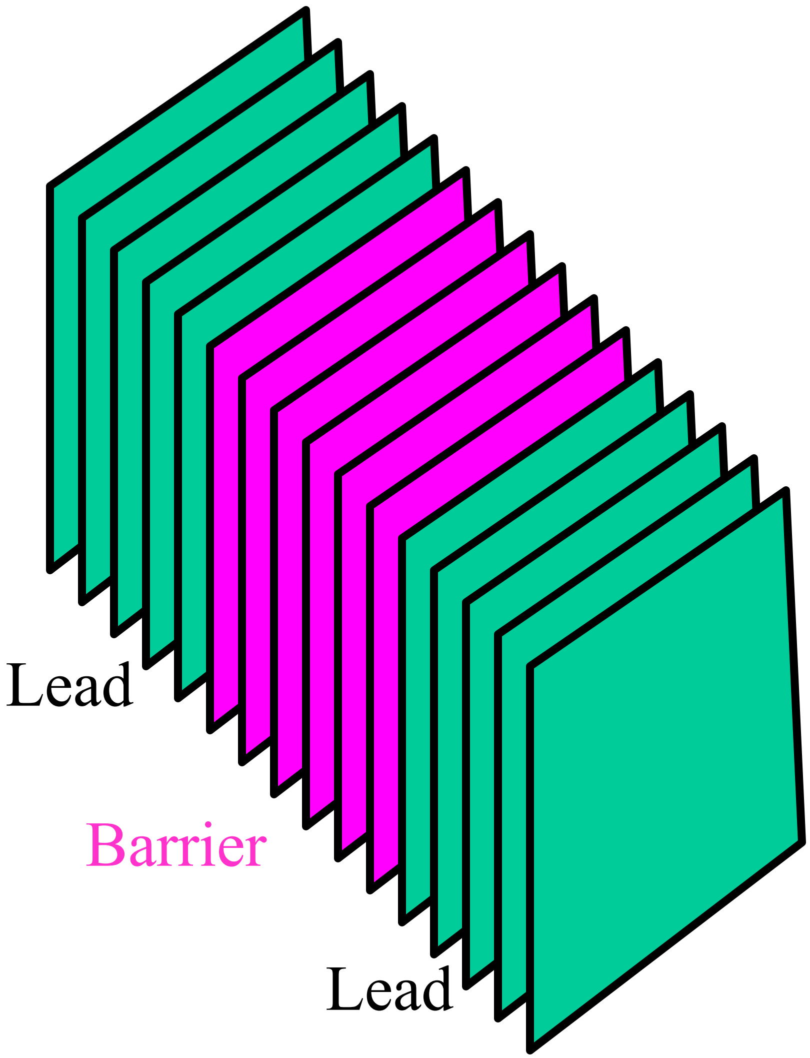
Theoretical Approaches (charge transport)

- Ohm's law: $R_n=\rho L/A$, holds for bulk materials
- Landauer approach: calculate resistance by determining the reflection and transmission coefficients for quasiparticles moving through the inhomogeneous device ($R_n=h/2e^2 * [1-T]/T$)
- Works well for ballistic metals, diffusive metals, and infinitesimally thin tunnel barriers ("delta function potentials").
- Real tunnel barriers have a finite thickness---the quasiparticle picture breaks down inside the insulating barrier; not clear that Landauer approach still holds.
- As the barrier thickness approaches the bulk limit, the transport crosses over to being thermally activated in an insulator and is no longer governed by tunneling.

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.

Our model



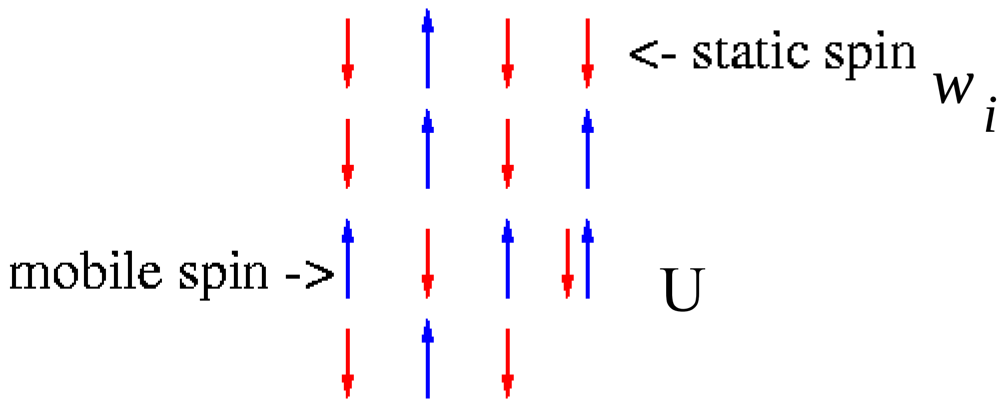
The metallic leads can be ballistic normal metals, mean-field theory ferromagnets, or BCS superconductors.

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

Scattering can also be included in the leads if desired, but we don’t do so here.

Spinless Falicov-Kimball Model

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

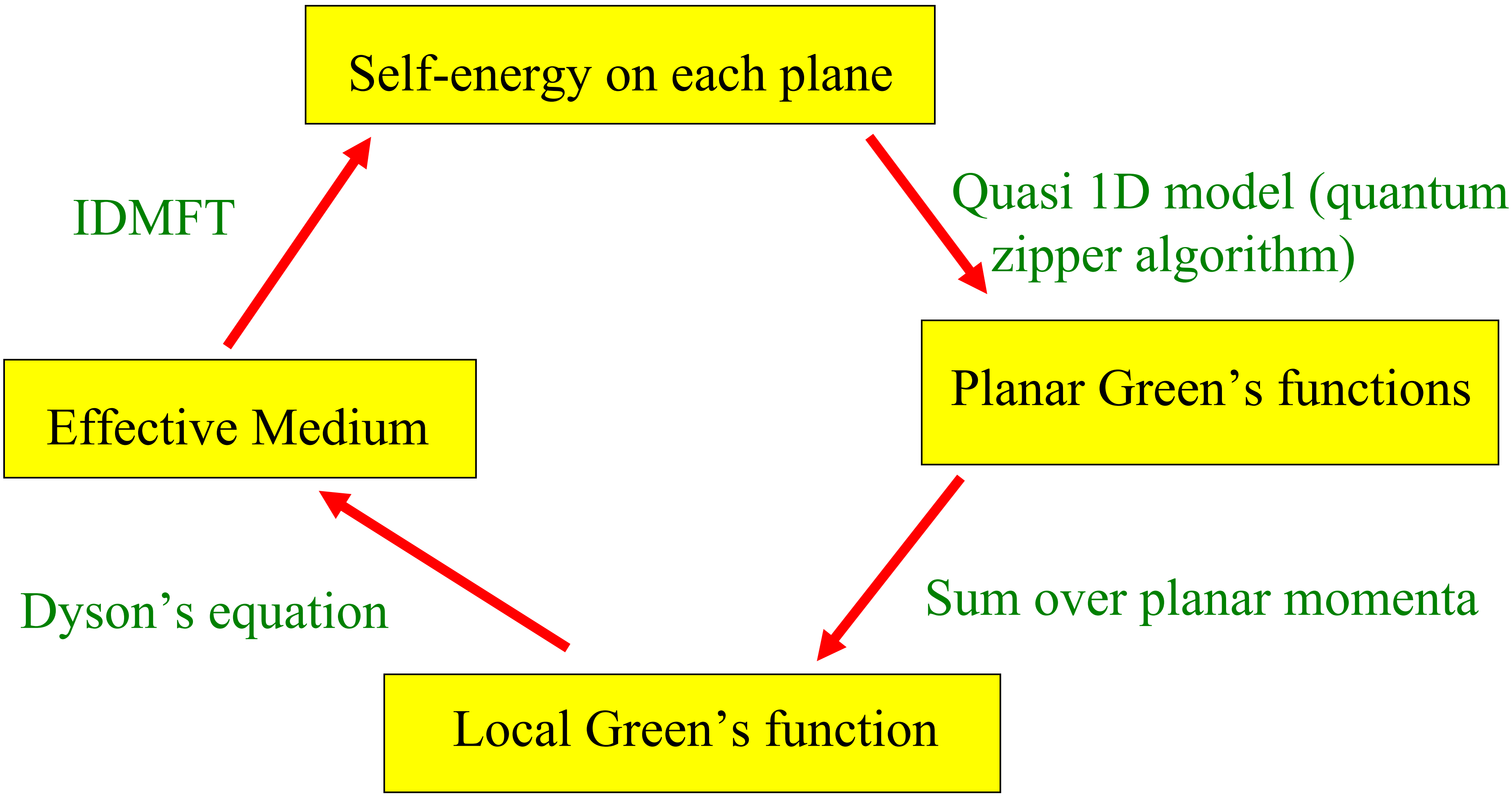


•**exactly solvable model** in the local approximation using dynamical mean field theory.

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

•***A self-consistent recursive Green's function approach solves the inhomogeneous many-body problem (Potthoff-Nolting algorithm).***

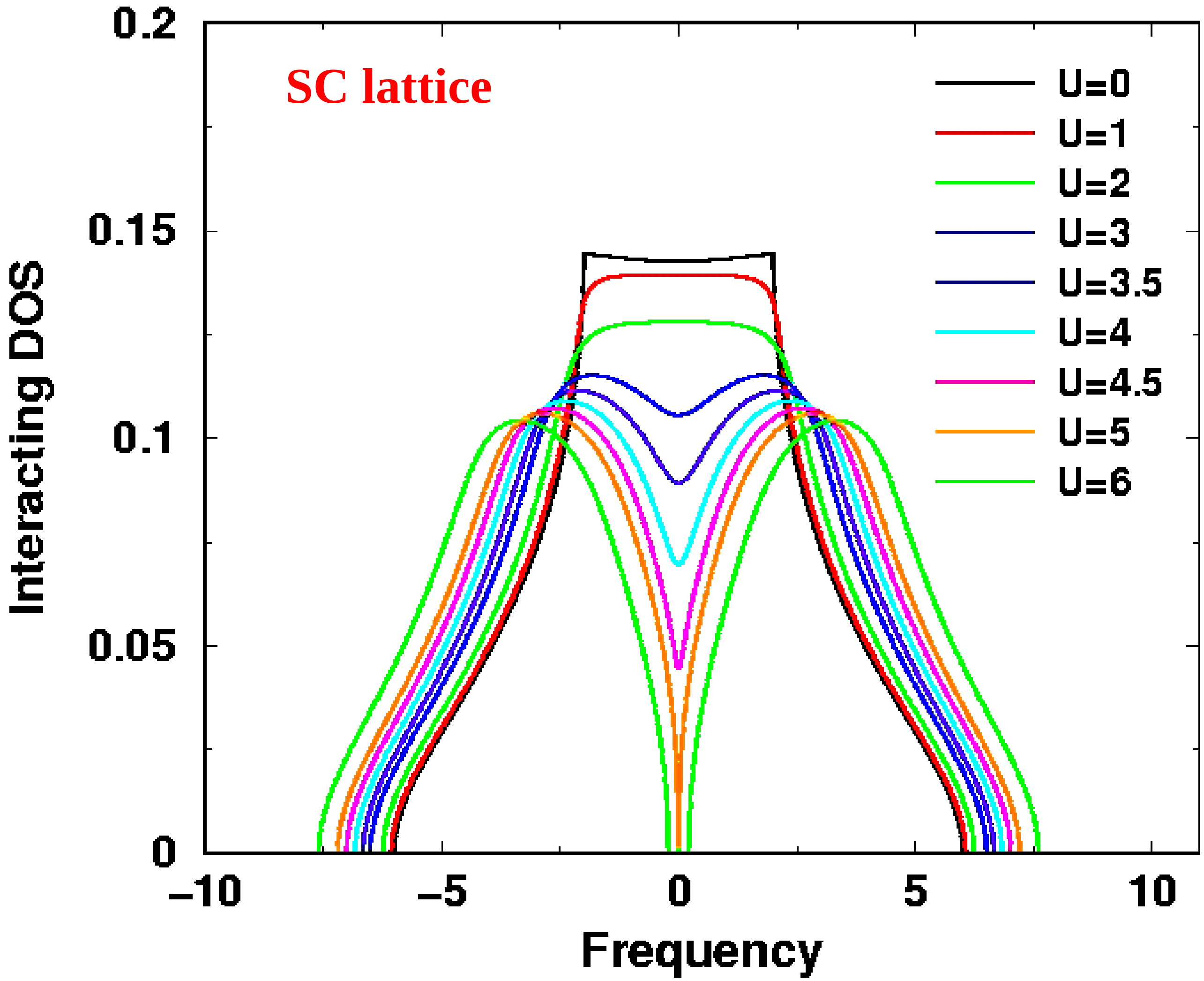
Computational Algorithm



Algorithm is iterated until a self-consistent solution is achieved

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

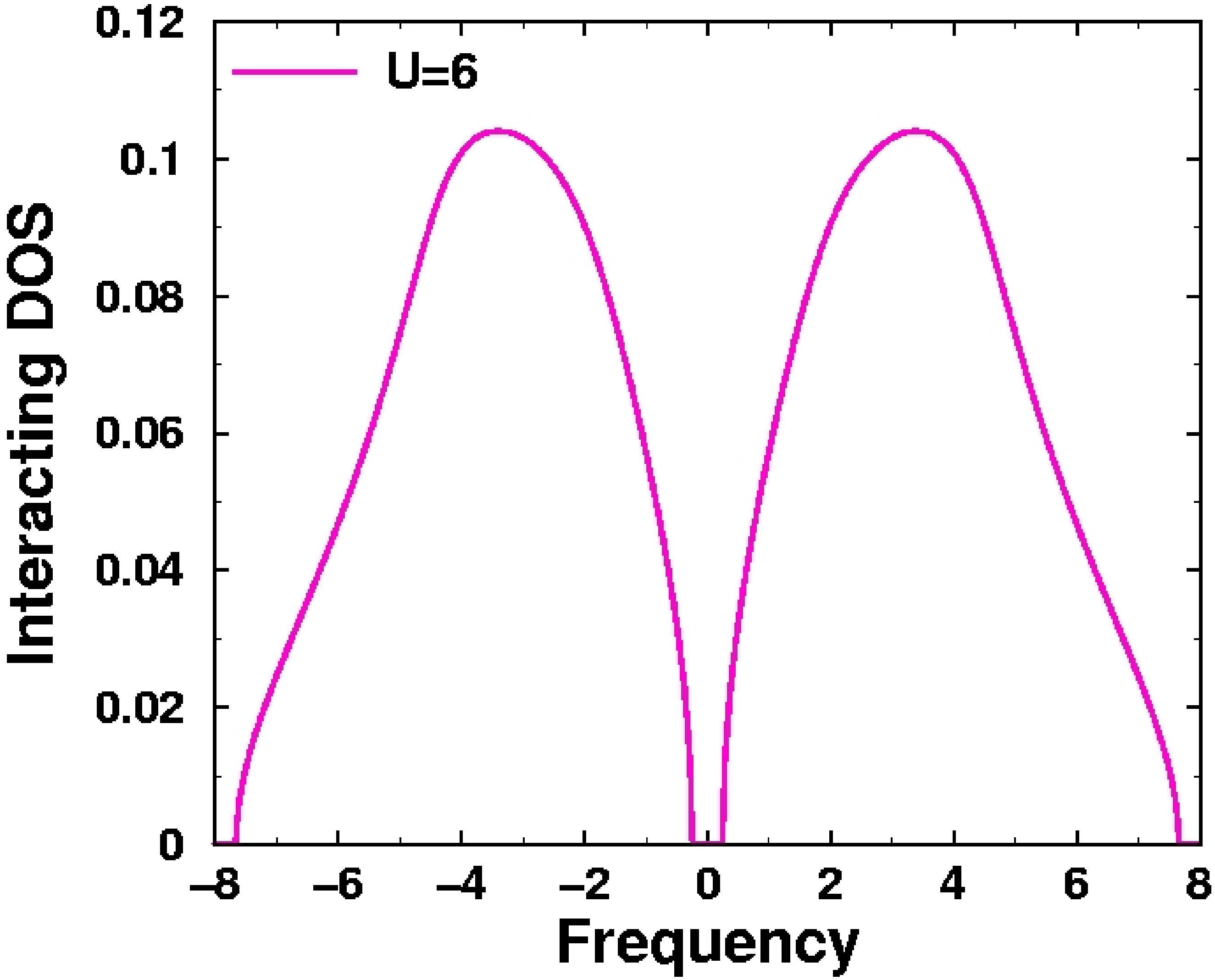
Metal-insulator transition (half-filling)



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.

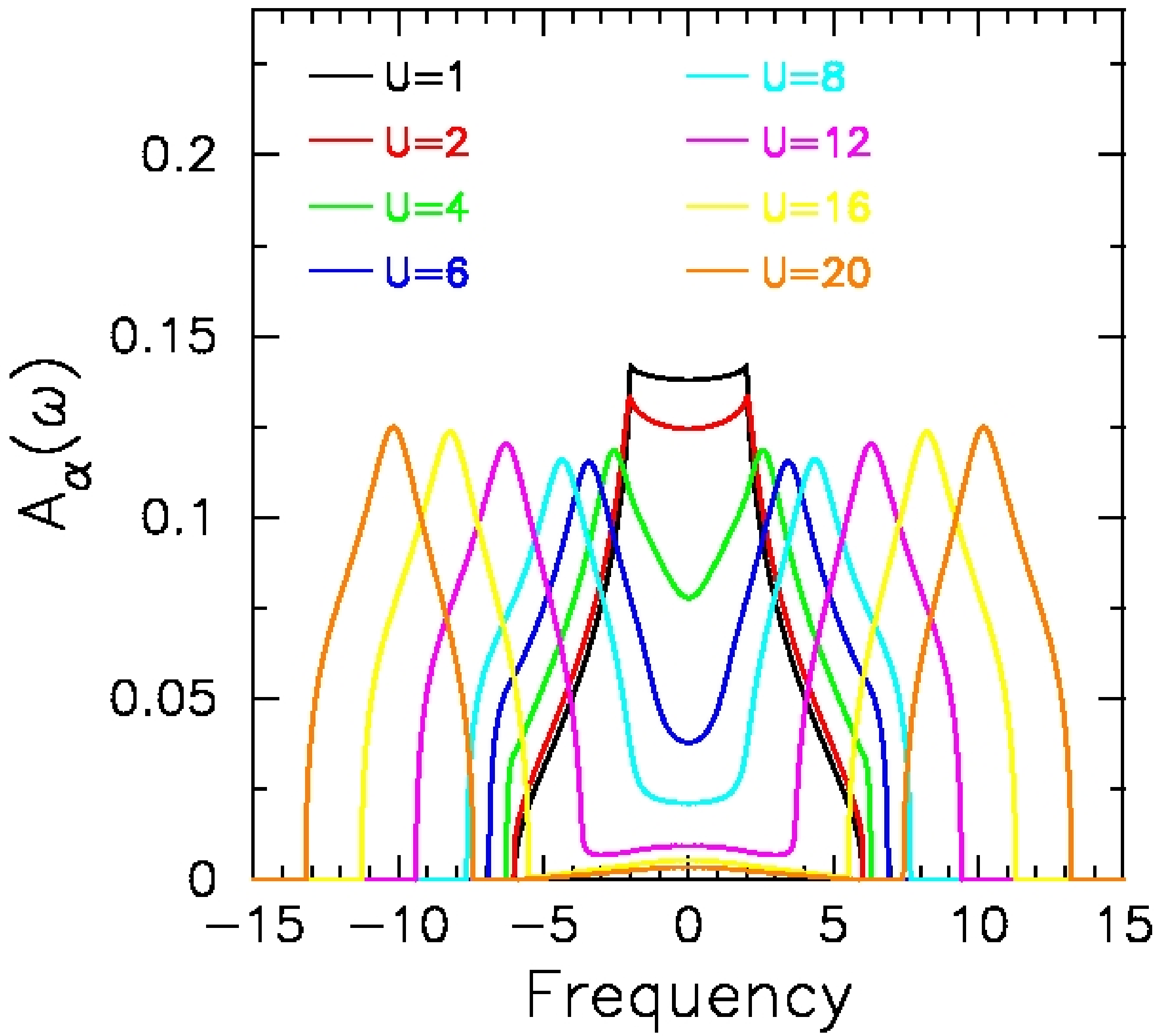
Near the MIT (U=6)



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.

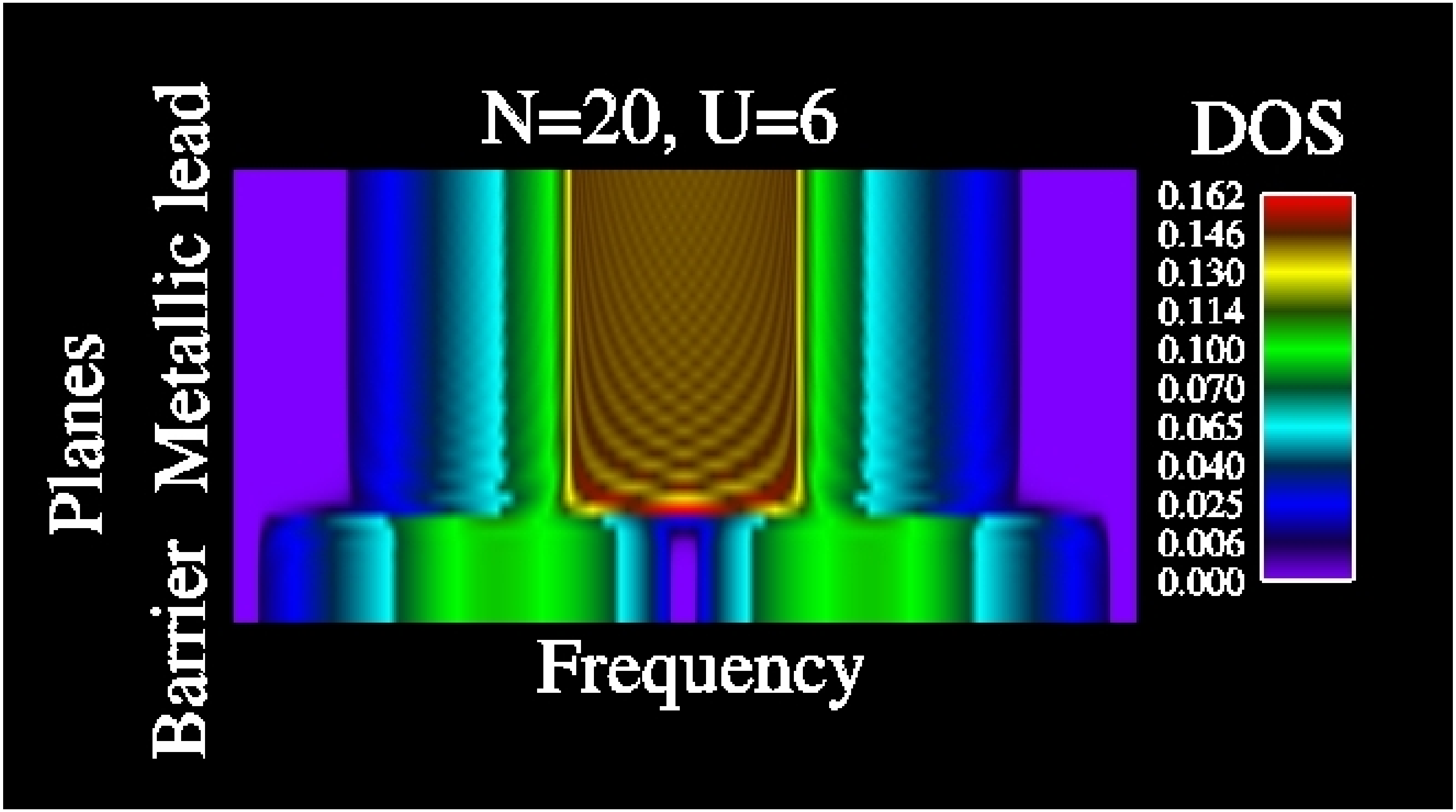
L=a (Single plane barrier)



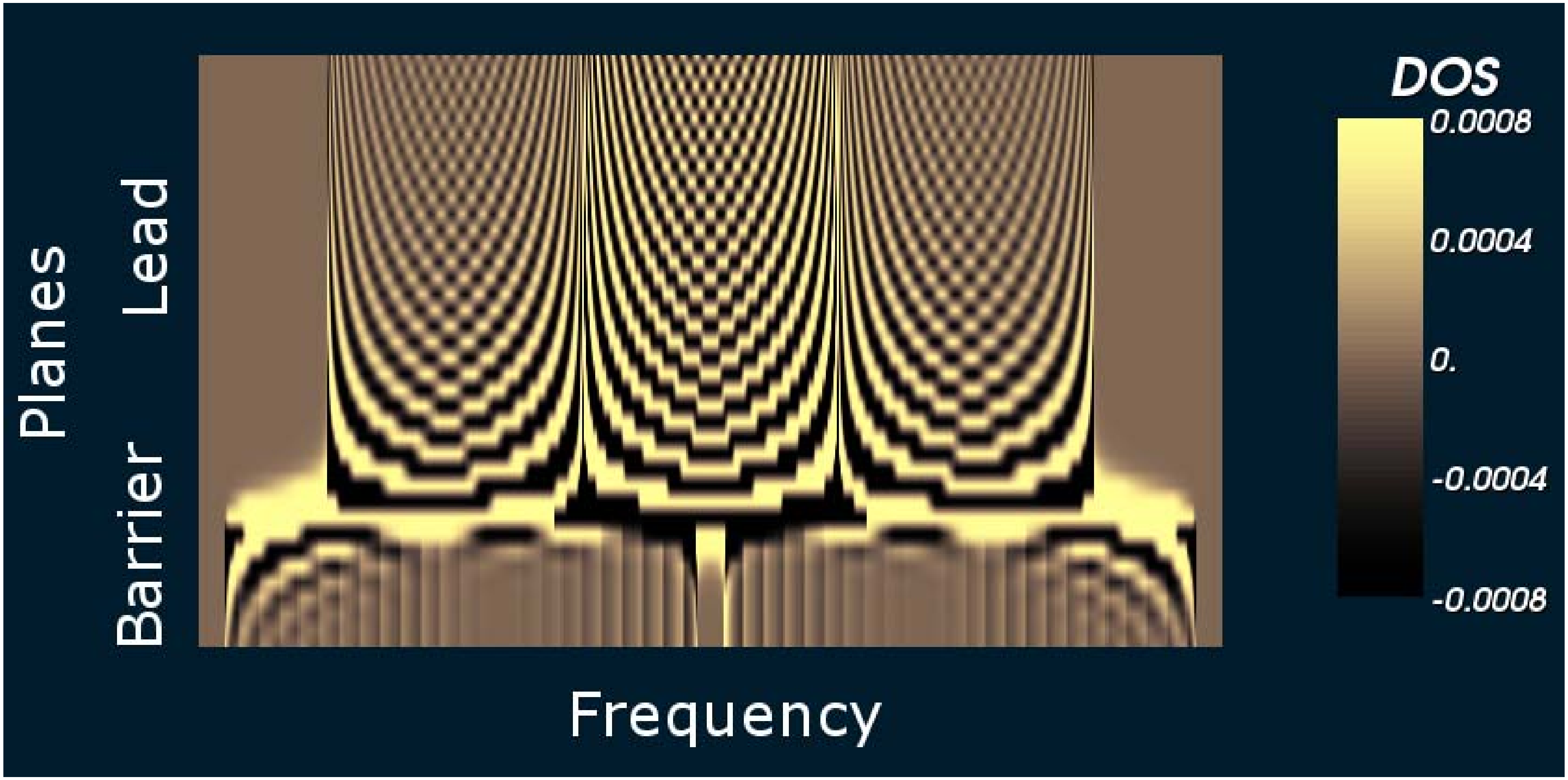
Local DOS on the central barrier plane. Note how the upper and lower Hubbard bands form for the Mott transition, but there is always substantial subgap DOS from the localized barrier states.

This DOS arises from quantum-mechanical tunneling and has a ***metallic shape.***

U=6 (small-gap insulator) DOS

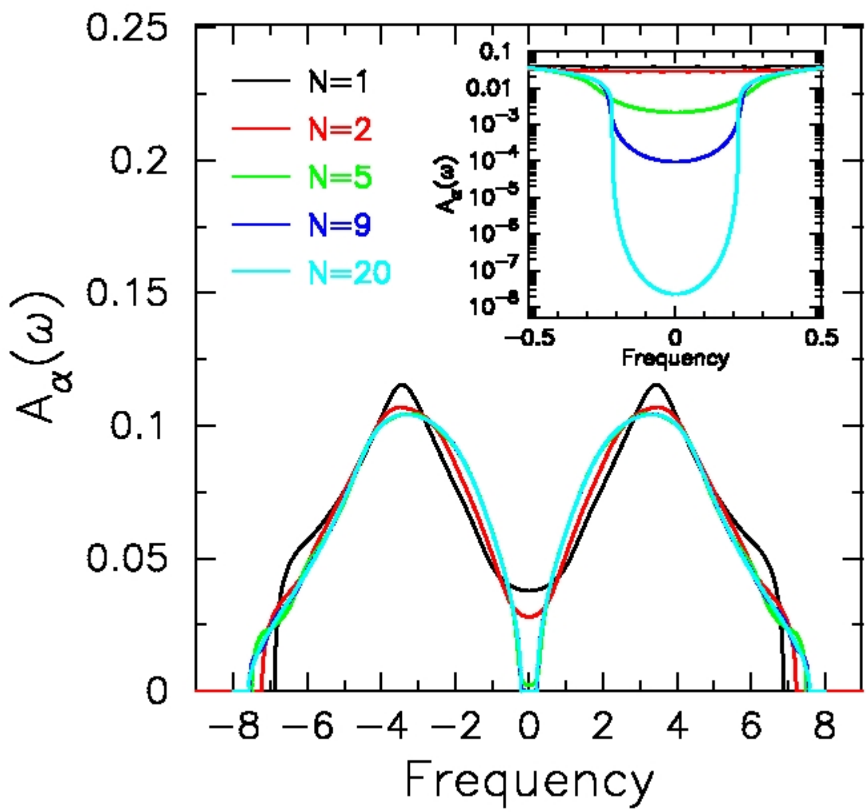


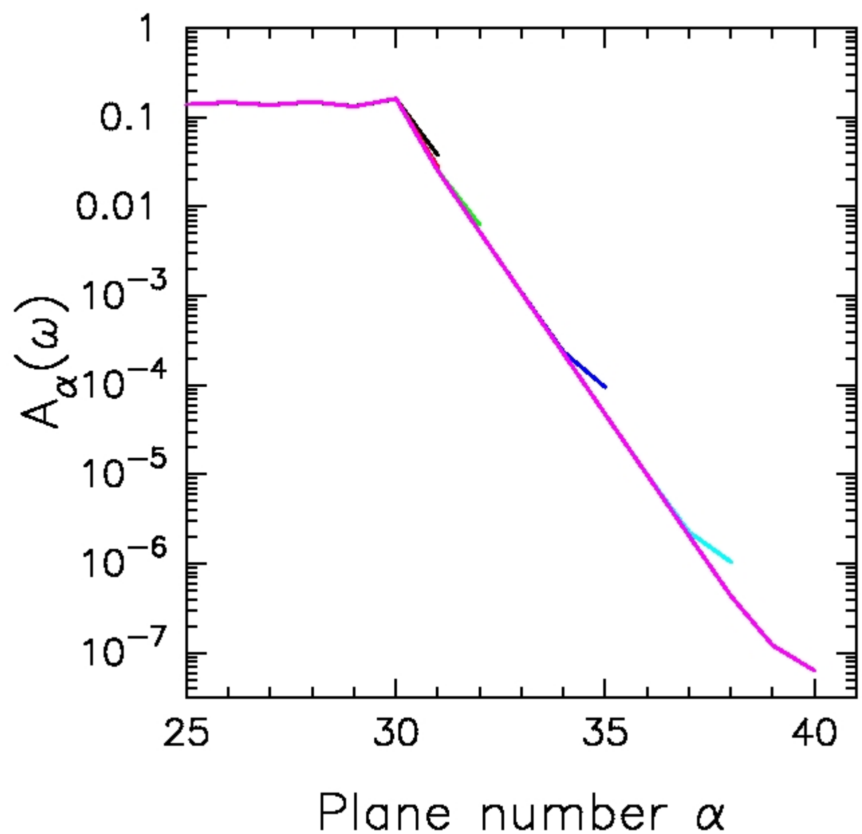
Friedel oscillations induced by interface



U=6 Correlated insulator

DOS has exponential tails, but never vanishes in the “gap”; the exponential decay has the same characteristic length for all barrier thicknesses.





Charge transport and the generalized Thouless energy …

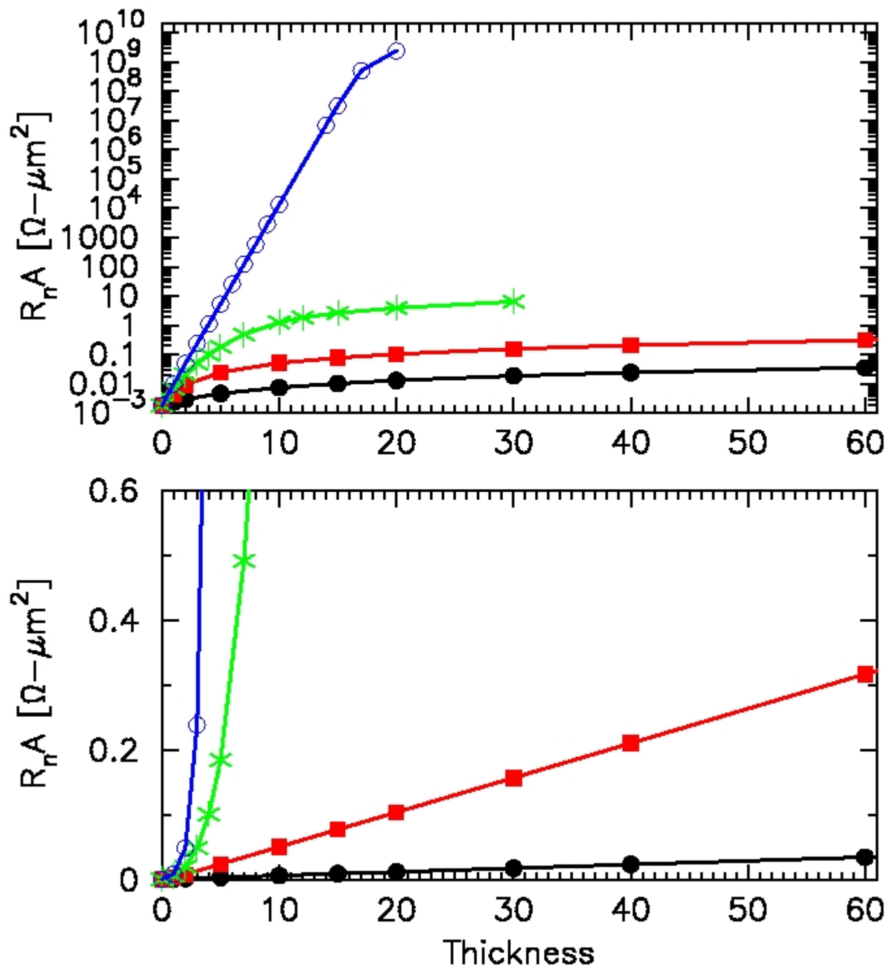
Junction resistance

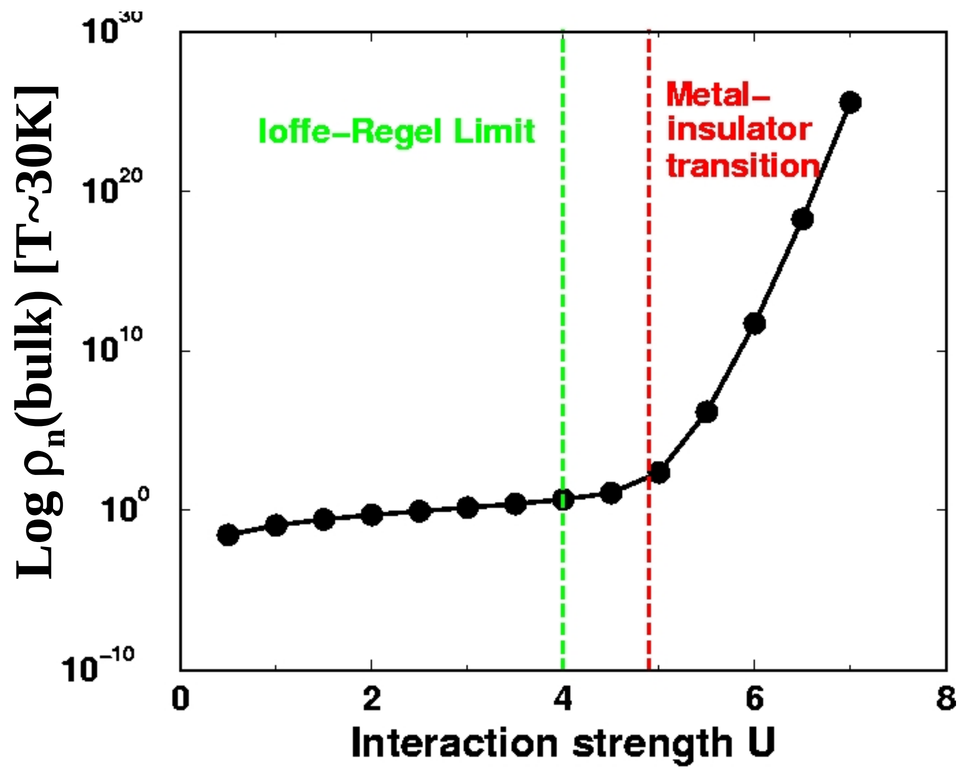
- The linear-response resistance can be calculated in equilibrium using a Kubo-Greenwood approach.
- We must work in real space because there is no translational symmetry.
- R_n is calculated by inverting the isothermal conductivity matrix and summing all matrix elements of the inverse.

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 $R_nA=a\sum_{ij}\sigma^{-1}_{ij}$

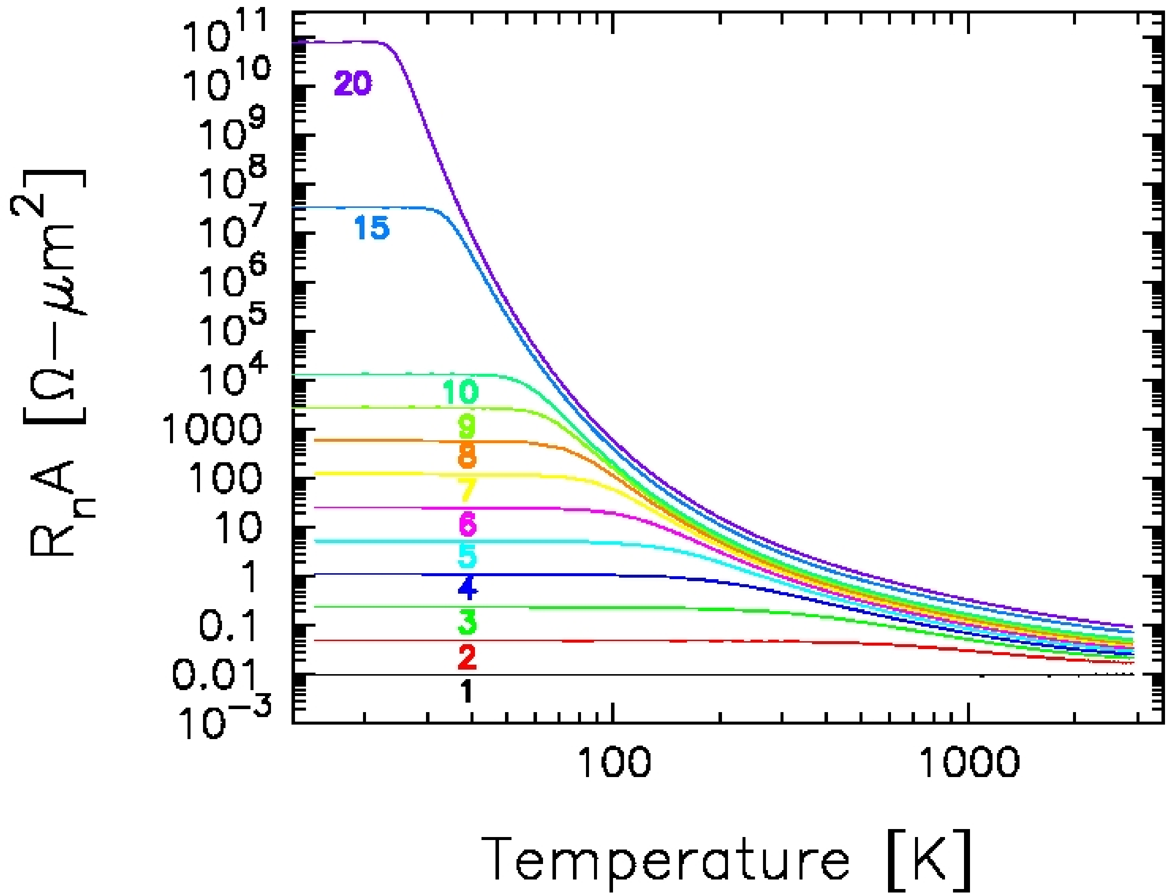
Resistance versus resistivity





Resistance for U=6 (correlated insulator)

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



Thouless energy

- The **Thouless energy** measures the quantum energy associated with the time that an electron spends inside the barrier region of width L (Energy extracted from the resistance).

$$E_{Th} = \hbar / t_{Dwell}$$

- A **unifying form** for the Thouless energy can be determined from the resistance of the barrier region and the electronic density of states:

$$E_{Th} = \frac{\hbar}{2e^2 \int d\omega N(\omega) \frac{-df(\omega)}{d\omega} R_N A L}$$

- This form produces both the **ballistic** $E_{Th} = \hbar v_F^N / \pi L$ and the **diffusive** $E_{Th} = \hbar D / L^2$ forms of the Thouless energy.

Thouless energy II

- The **resistance** can be considered as the **ratio** of the Thouless energy to the quantum-mechanical level spacing Δ_E (with R_Q=h/2e^2 the quantum unit of resistance)

$$R_n = R_Q \frac{\Delta_E}{2\pi E_{Th}}$$

- The inverse of the level spacing is related to the density of states of the barrier via

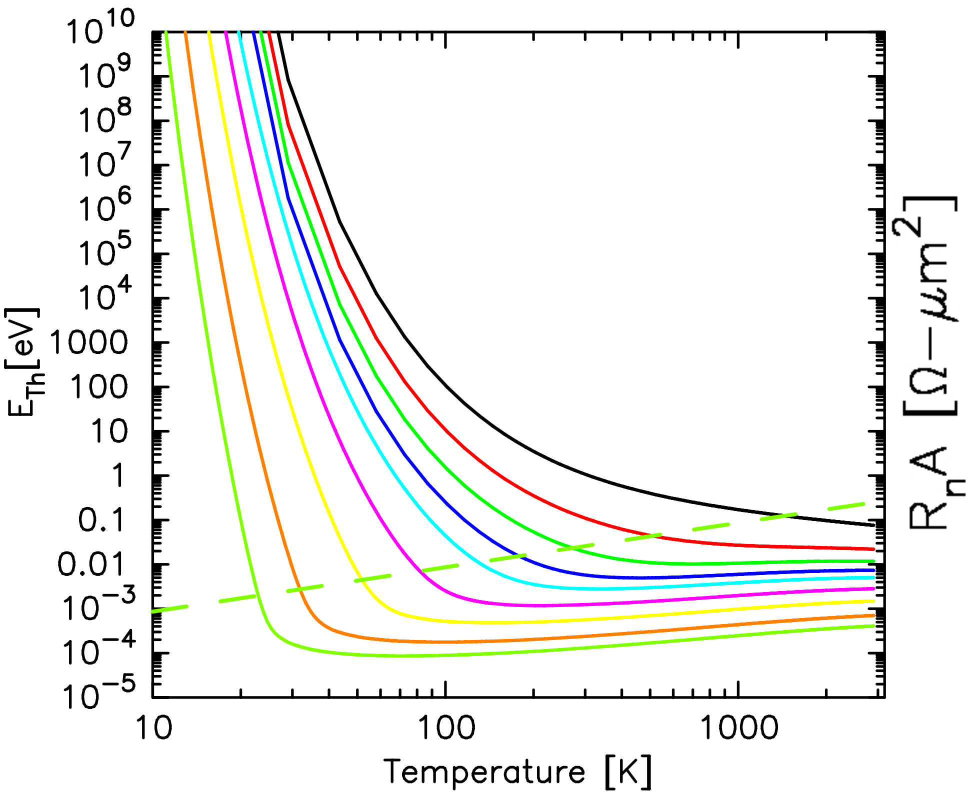
$$\Delta_E^{-1} = VN(\mu)$$

- Generalizing the above relation to an insulator by

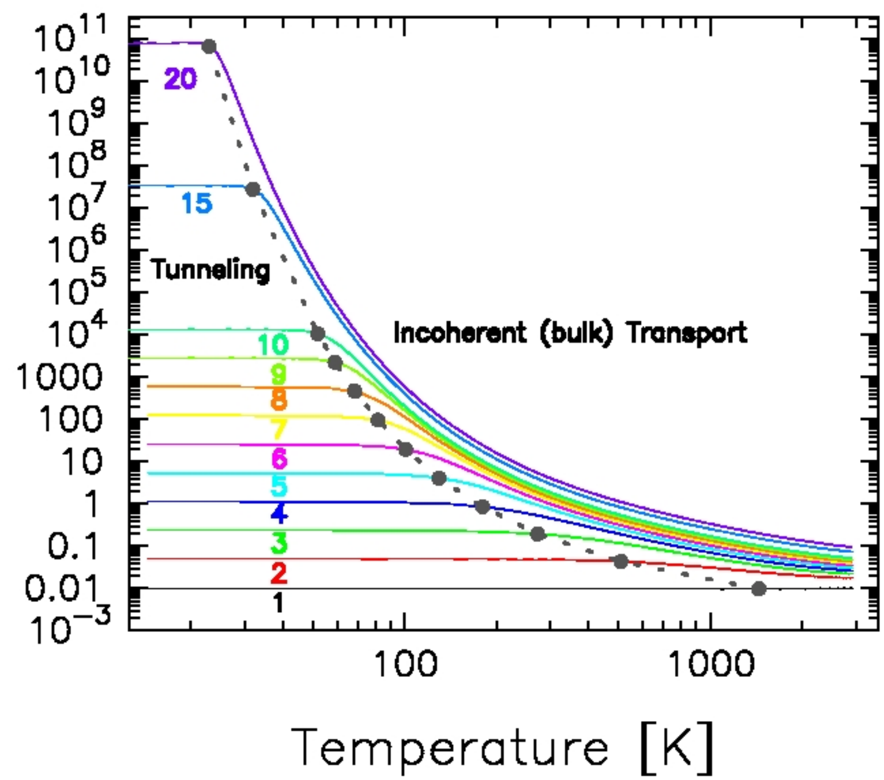
$$\Delta_E^{-1} = AL\int d\omega N(\omega)\left[-\frac{df(\omega)}{d\omega}\right]$$

gives the general form for the Thouless energy.

Temperature dependence



U=6 FK model



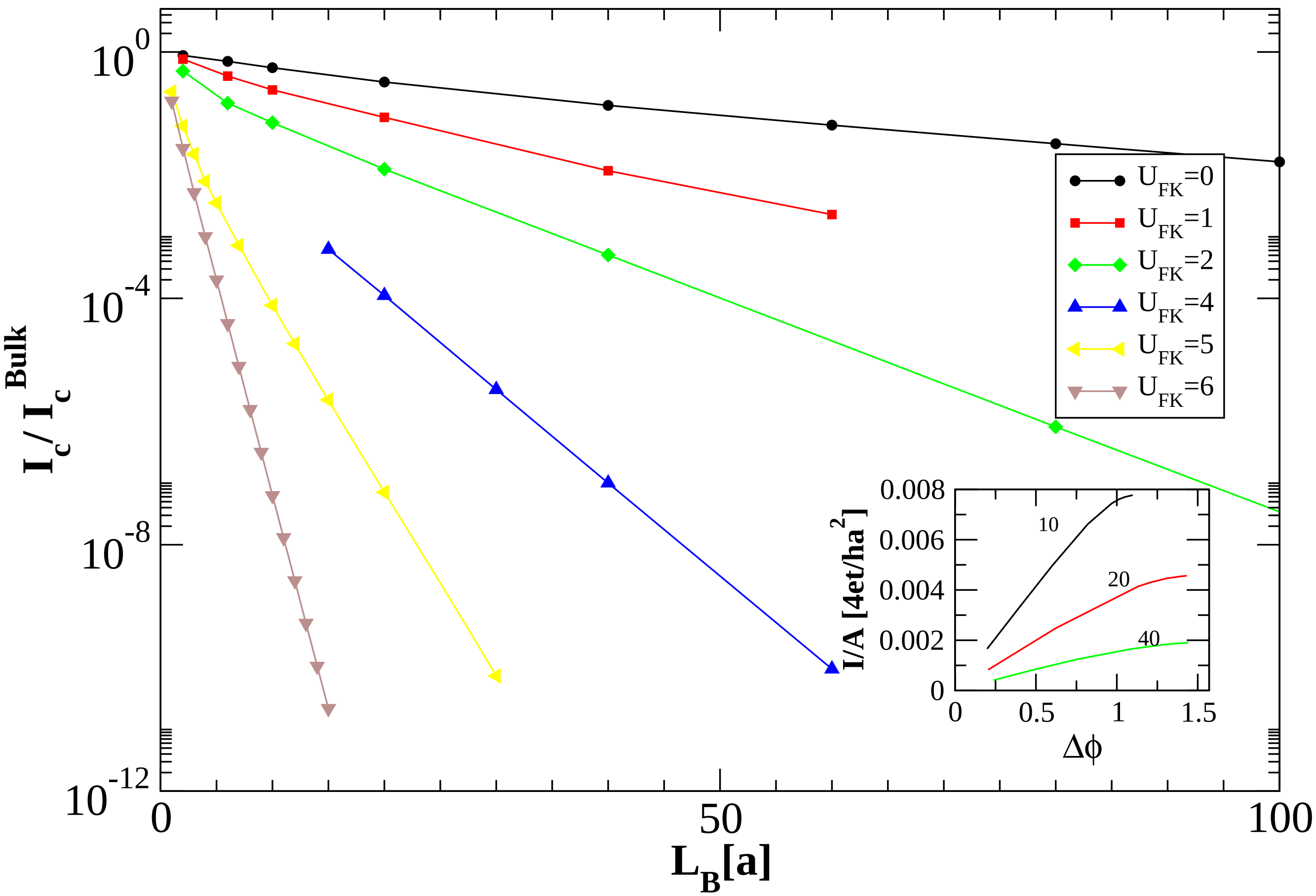
The Thouless energy determines the transition from tunneling to incoherent transport as a function of temperature!

Note that the crossover temperature is not simply related to the energy gap!

Josephson junctions, figure-of-merit, and the generalized Thouless energy …

Critical current density

The current throug[h] the junction is increased as a phas[e] gradient is placed over the leads. The current increases t[o] a maximal value, called the critical current of the junction. Here we show the critical current versus the thickness of the barrier.



Switching time of a junction

In a Josephson junction, the integral of a voltage pulse over time is equal to a flux quantum. Hence, increasing the magnitude of the voltage pulse, I_cR_n, will minimize the width of the pulse, and hence produce the fastest switching speed.

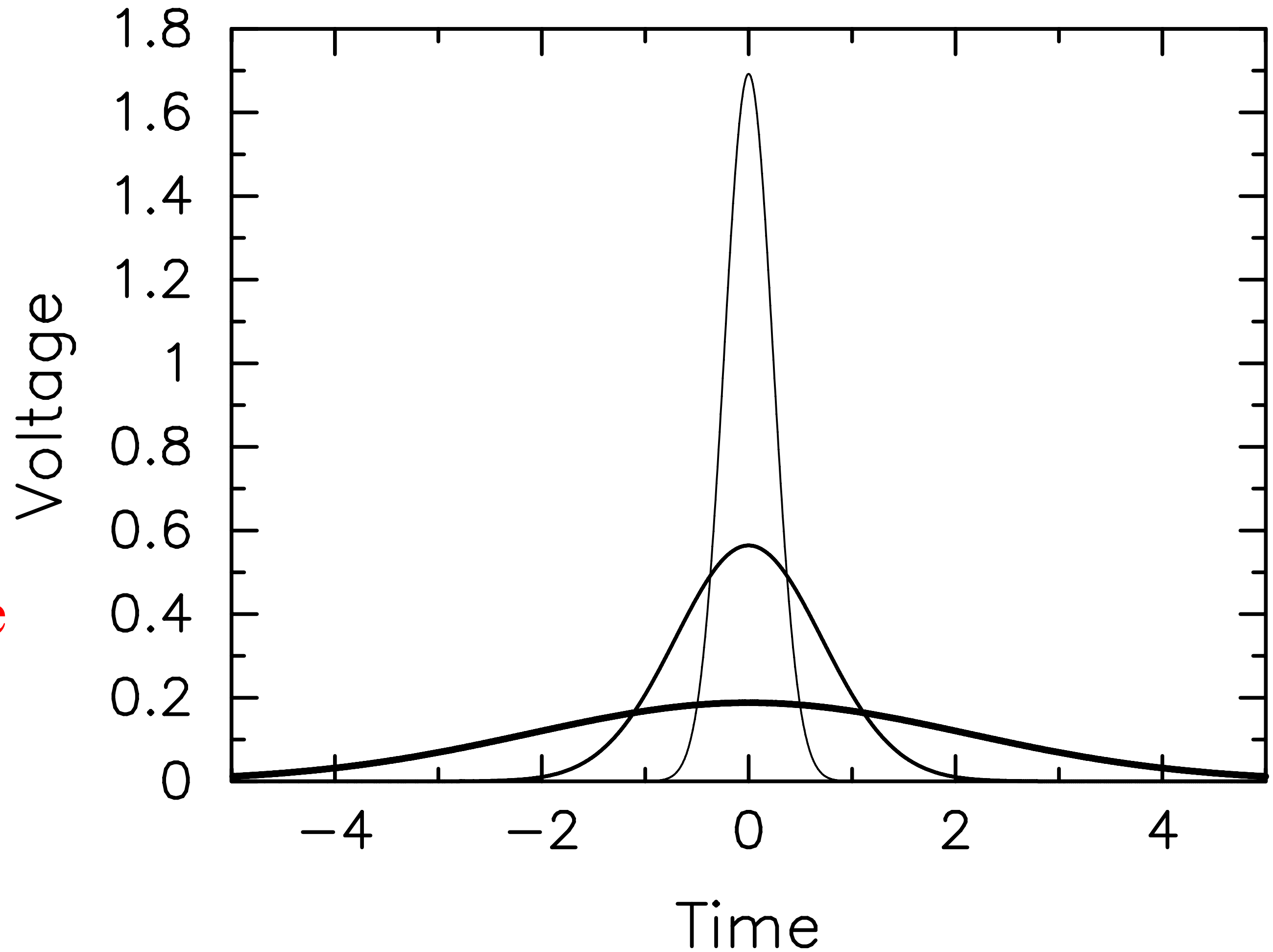
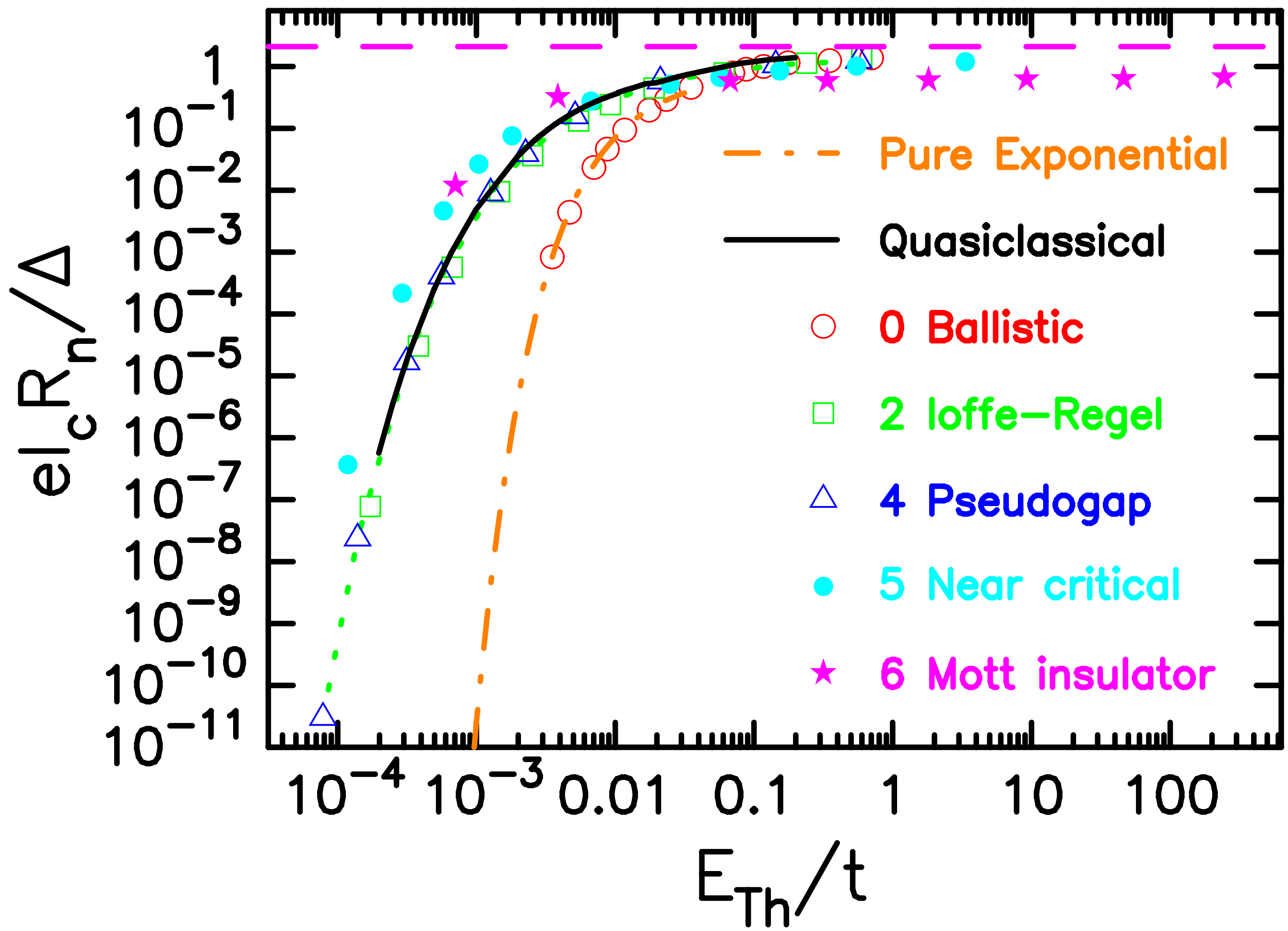


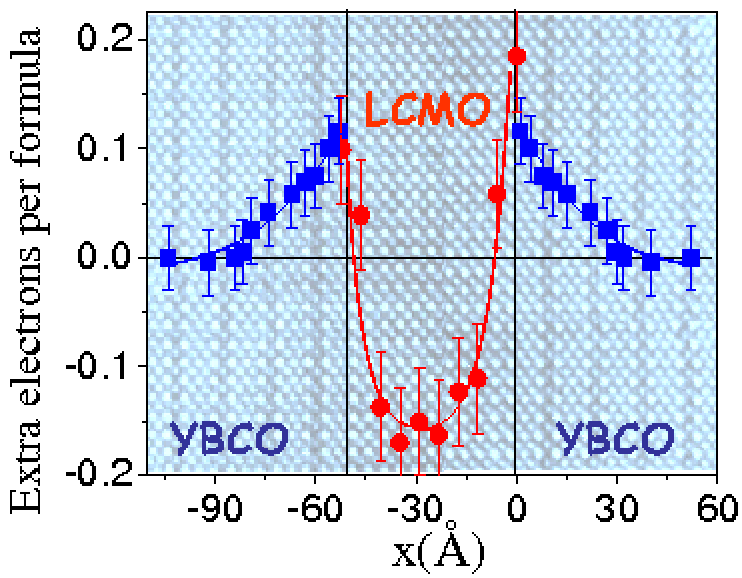
Figure of Merit

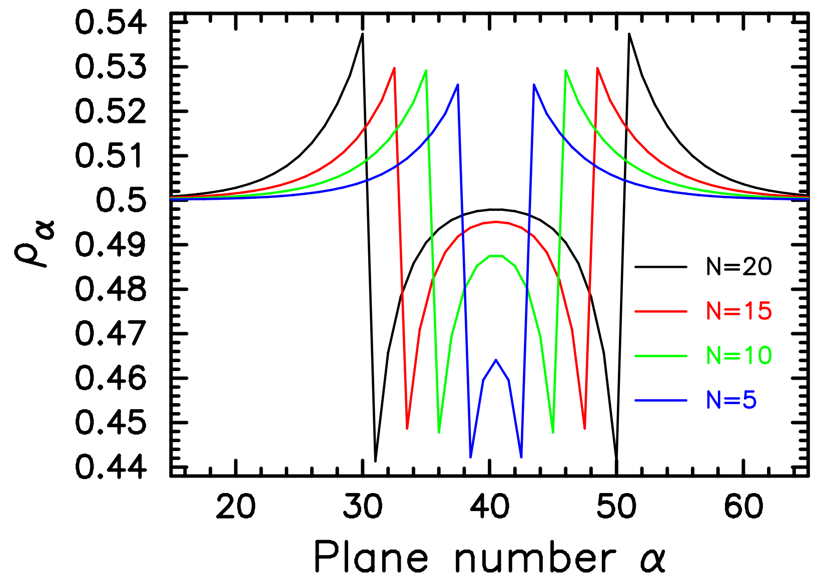
The Thouless energy determines the figure of merit once it becomes the **smallest energy scale** in the problem. The quasiclassical approach seems to hold **all they way up to the MIT**. Beyond that, the behavior appears to become **nonuniversal**, but with similar qualitative features.



Particle-hole asymmetry is necessary for thermoelectric devices …

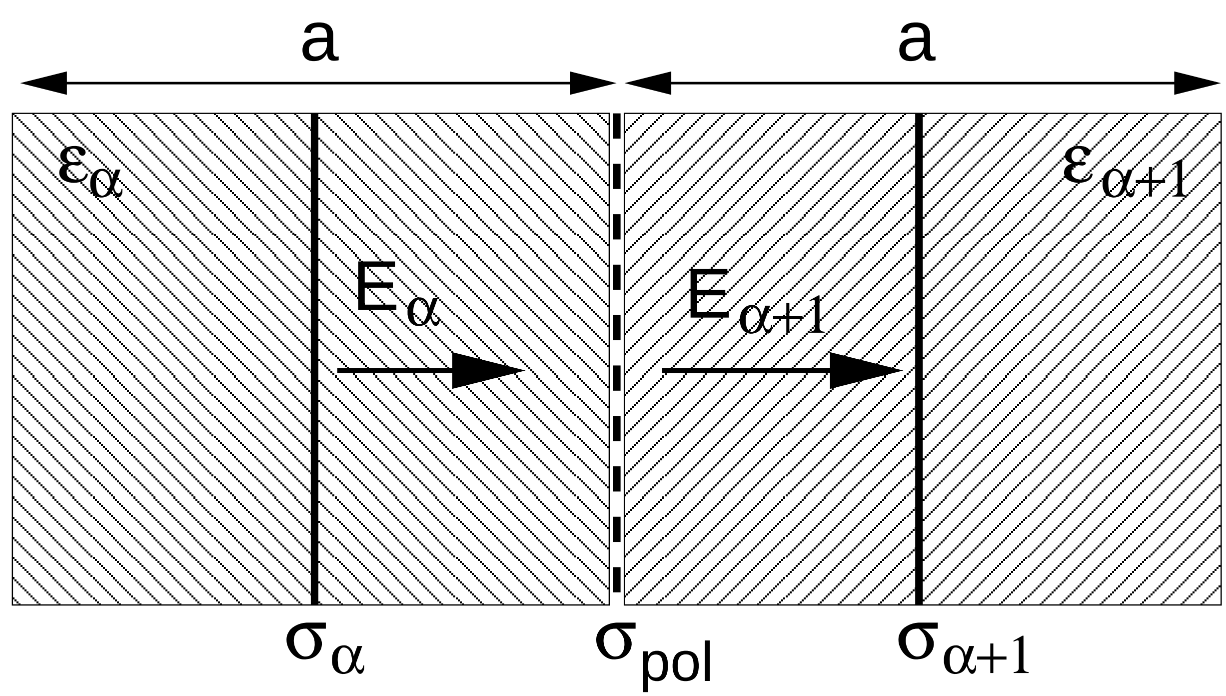
Electronic charge reconstruction





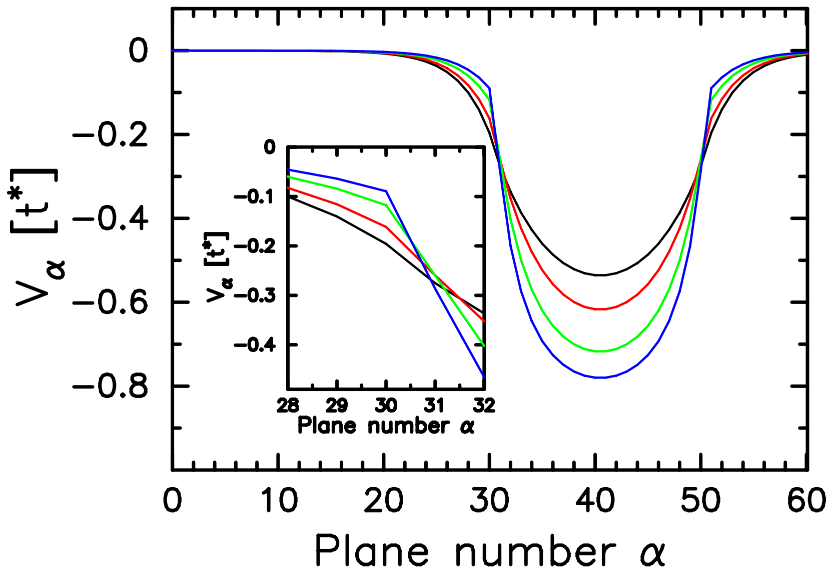
Using a scanning transmission electron microscope with electron energy-loss spectroscopy, one can directly measure the electronic charge at each plane of a strongly correlated multilayered nanostructure. Left are experimental results by Varela et al. on YBCO/LCMO heterostructures, right is a simple theory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

Theoretical treatment

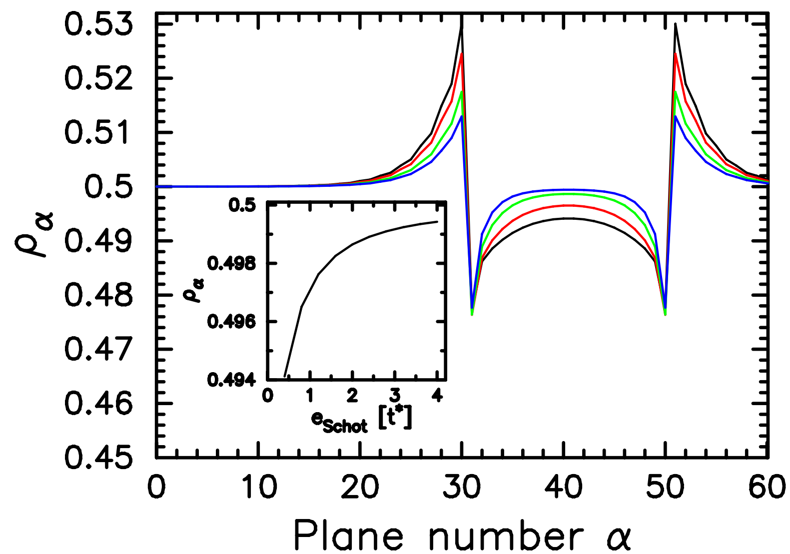


We employ a semiclassical treatment to handle the electronic charge reconstruction. We allow charge to be rearranged on different planes, as determined by the electrochemical potential at a given plane site, and then determine the classical Coulomb potential from planes of net charge, with dielectric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

Coulomb potential

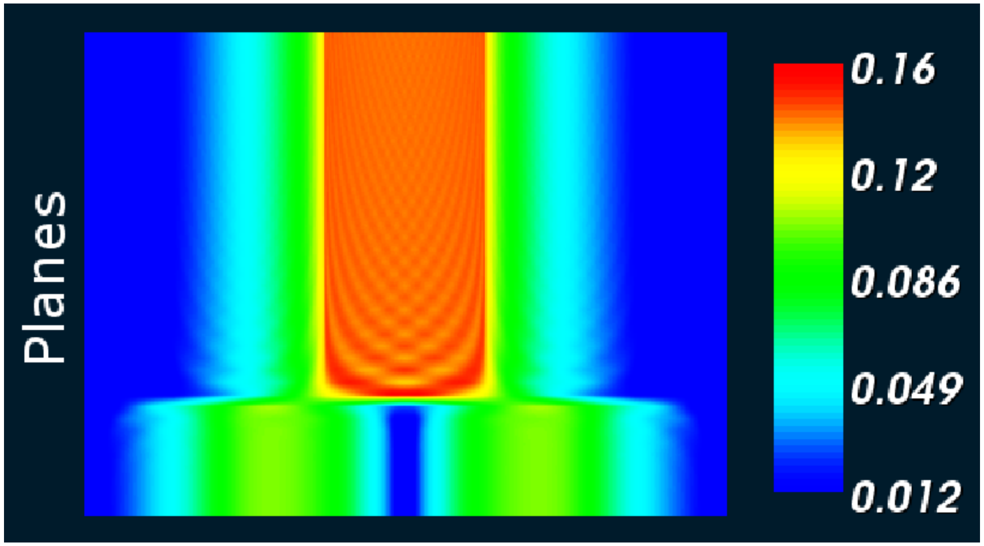


The Coulomb potential develops a kink at locations where the dielectric constant changes (i.e. at the interfaces), and it goes to zero far from the interface due to overall conservation of charge.

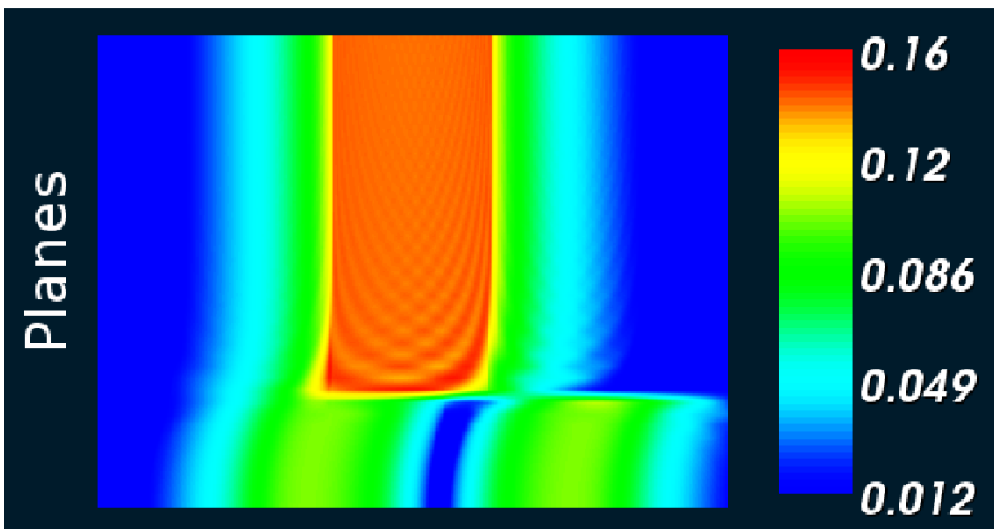


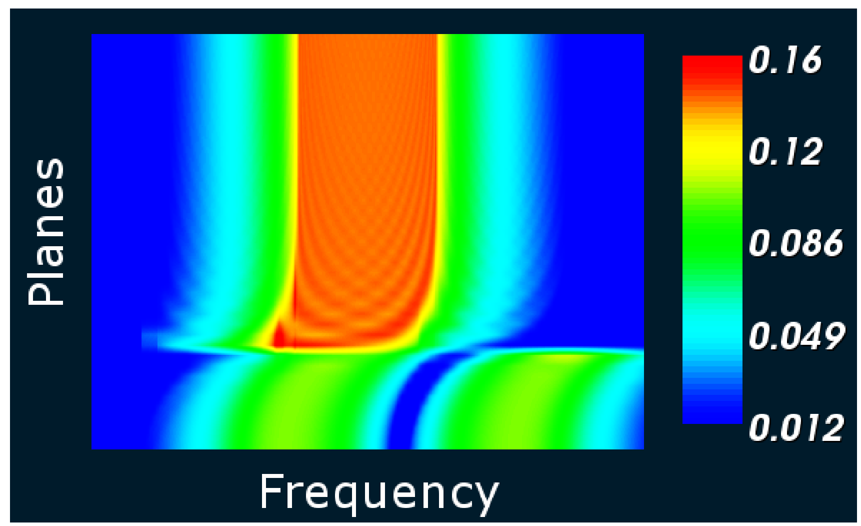
As the screening length decreases, the total charge that is rearranged gets smaller for a fixed chemical potential mismatch of the bulk materials.

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

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

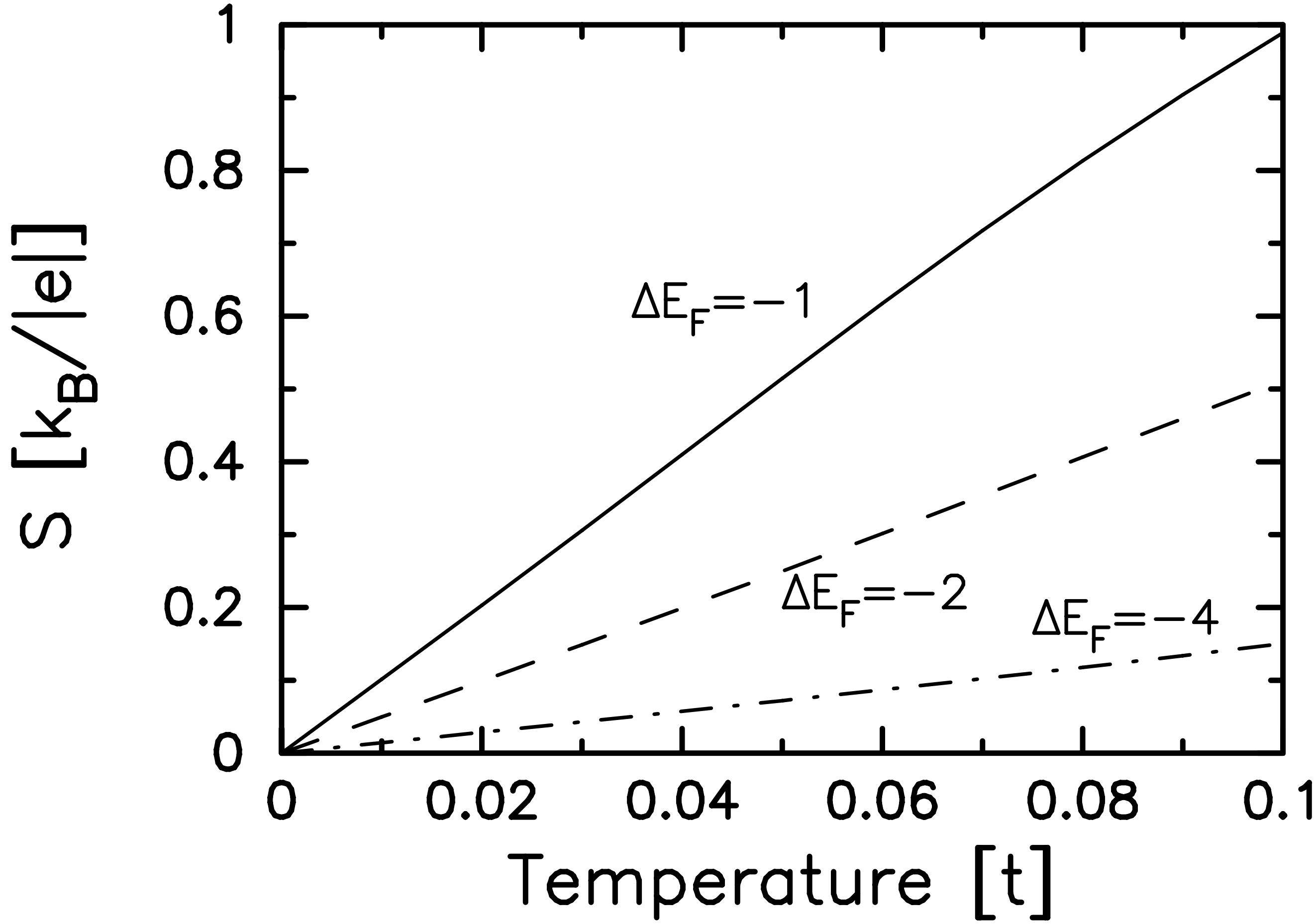
Hence, $\Delta T = -\sum_{ij} M^{-1}{}_{ij} j_Q$, $\Delta V = -a\sum_{ij}[(L^{11})^{-1}L^{12}M^{-1}]_{ij} j_Q$, and the Seebeck coefficient is

$$S = \Delta V/\Delta T = a\sum_{ij}[(L^{11})^{-1}L^{12}M^{-1}]_{ij} / \sum_{ij} M^{-1}{}_{ij}$$

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!

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!



Conclusions

In this talk I have covered a number of topics in strongly correlated nanostructures. These included the following: (i) DOS and charge transport in the particle-hole symmetric case, when the barrier is tuned through the Mott transition; (ii) a description of transport, including the tunneling to Ohmic crossover, via a generalized Thouless energy; (iii) an application to Josephson junctions and the figure-of-merit; (iv) electronic charge reconstruction, and how to self-consistently determine the screened dipole layers that lead to Schottky-like barriers; and (v) thermal transport and the Seebeck effect.

The interplay between inhomogeneity and electron-electron correlations leads to interesting new phenomena, that could have wide-ranging applications.