Thermal transport in strongly correlated nanostructures

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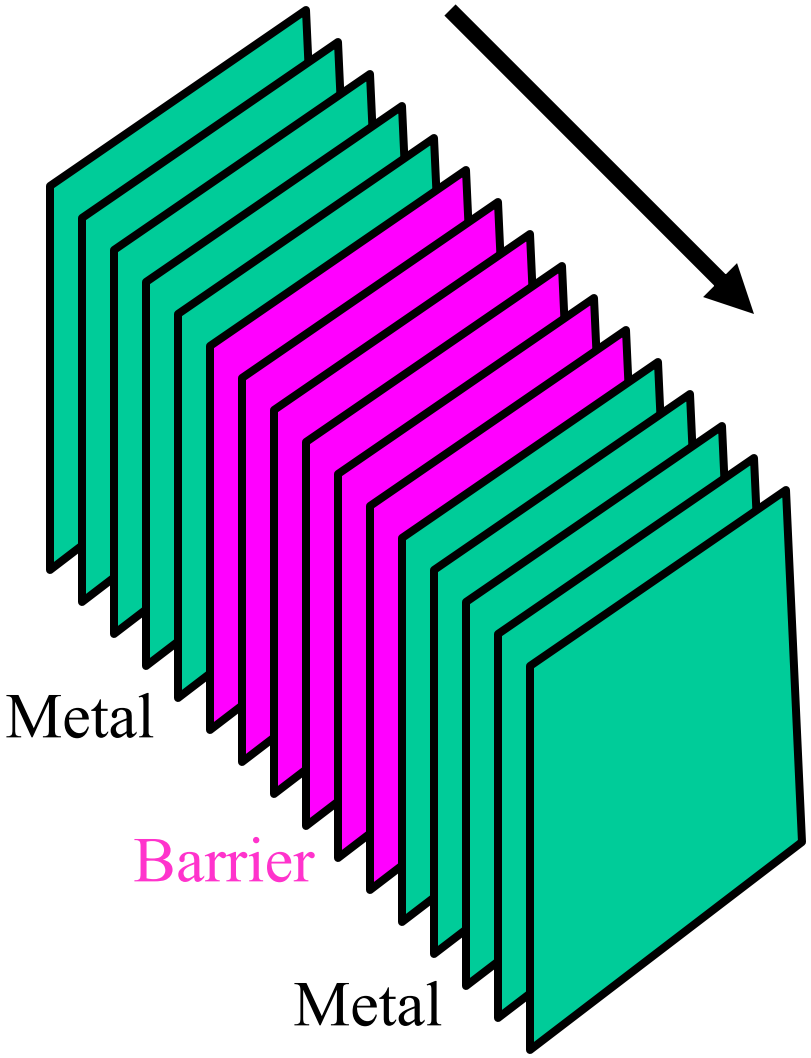
Multilayered nanostructures as devices



- 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

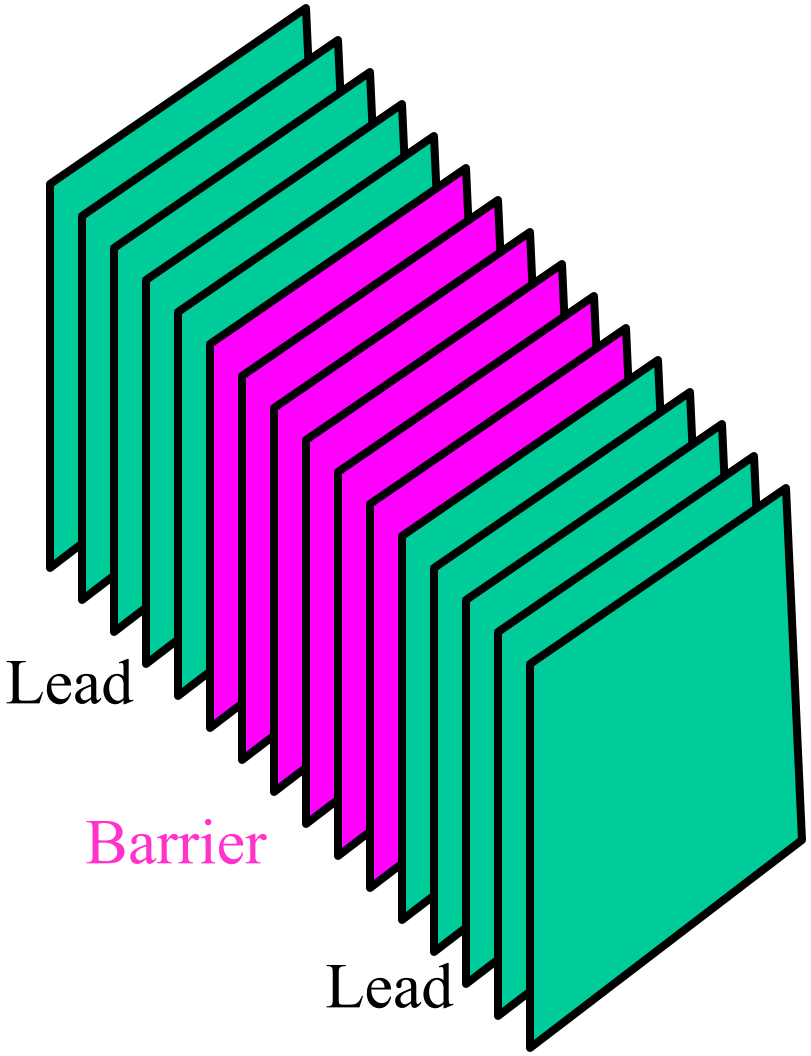
Theoretical Approaches (charge transport)

- 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 handle all of these different kinds of transport.

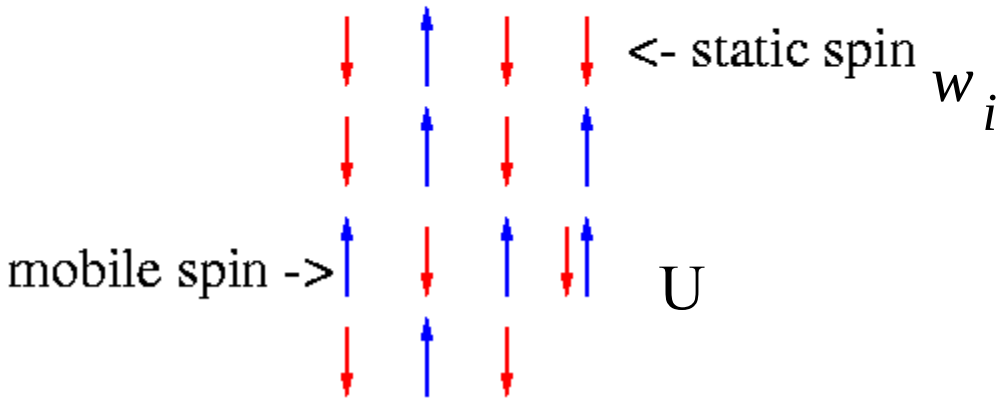
Our model



- 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" (Falicov-Kimball model)
- Scattering can also be included in the leads if desired, but we don't do so here.

Spinless Falicov-Kimball Model

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

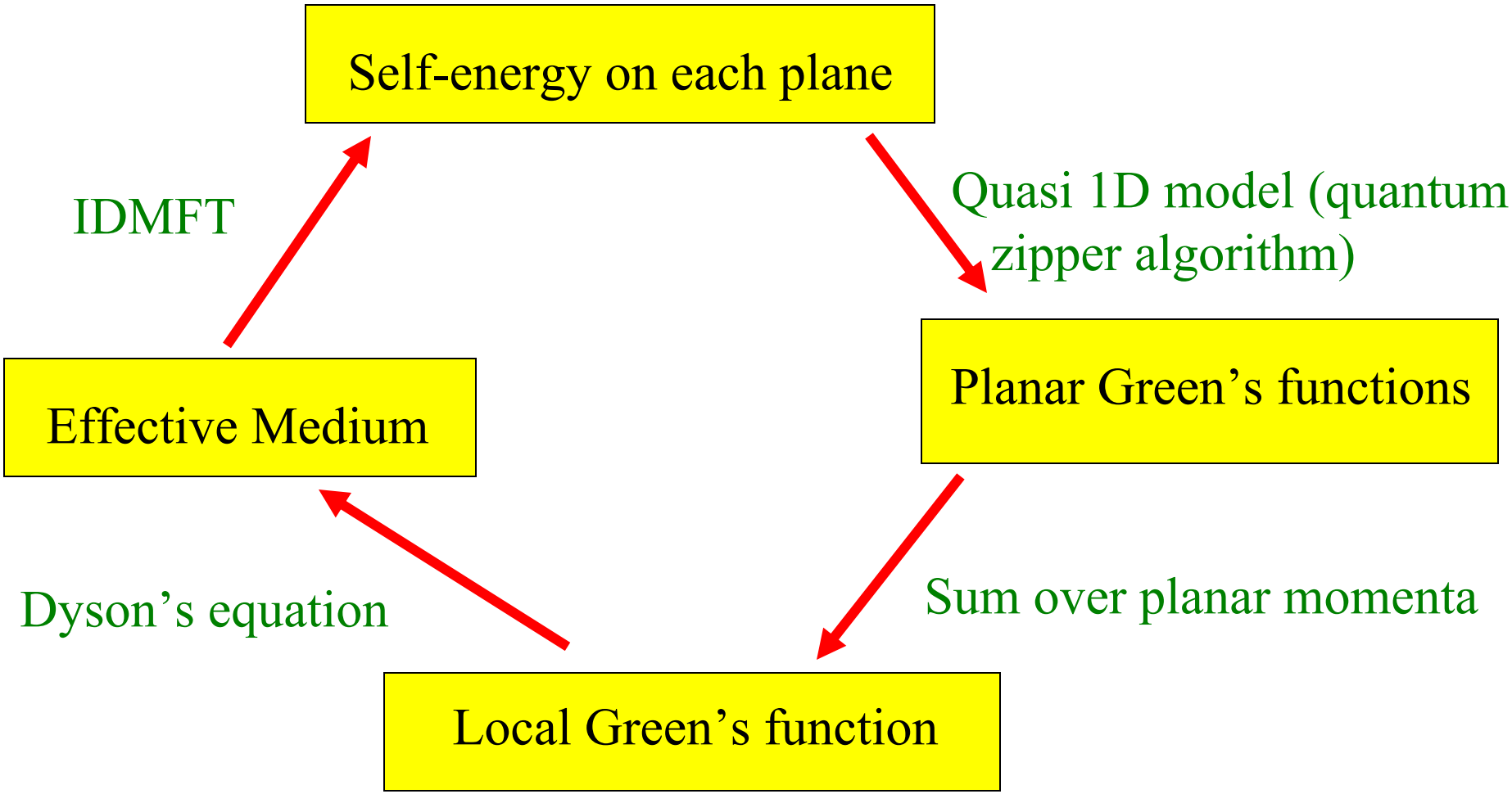


- **exactly solvable model** in the local approximation using dynamical mean field theory.

- 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).***

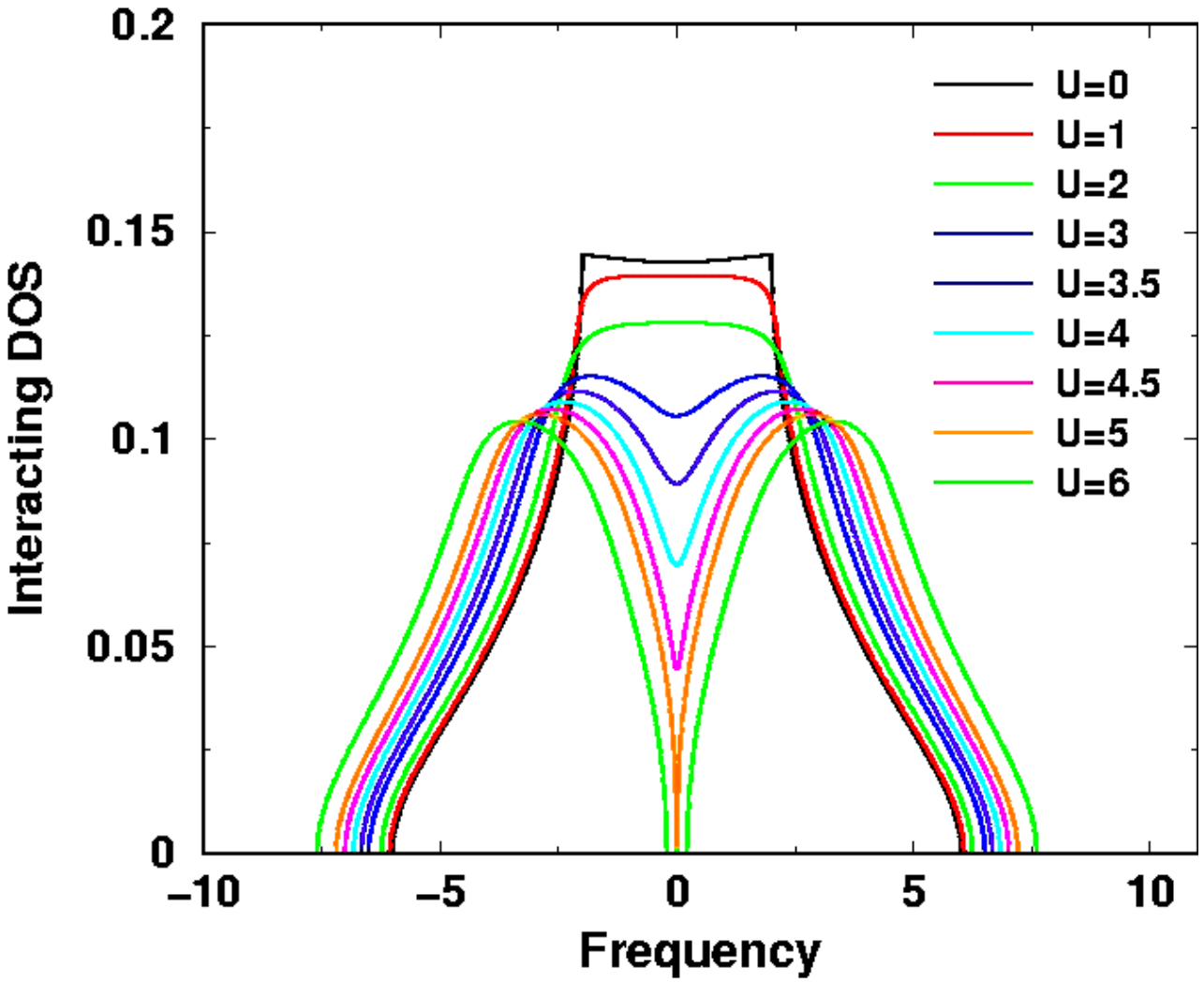
Computational Algorithm



Algorithm is iterated until a self-consistent solution is achieved

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

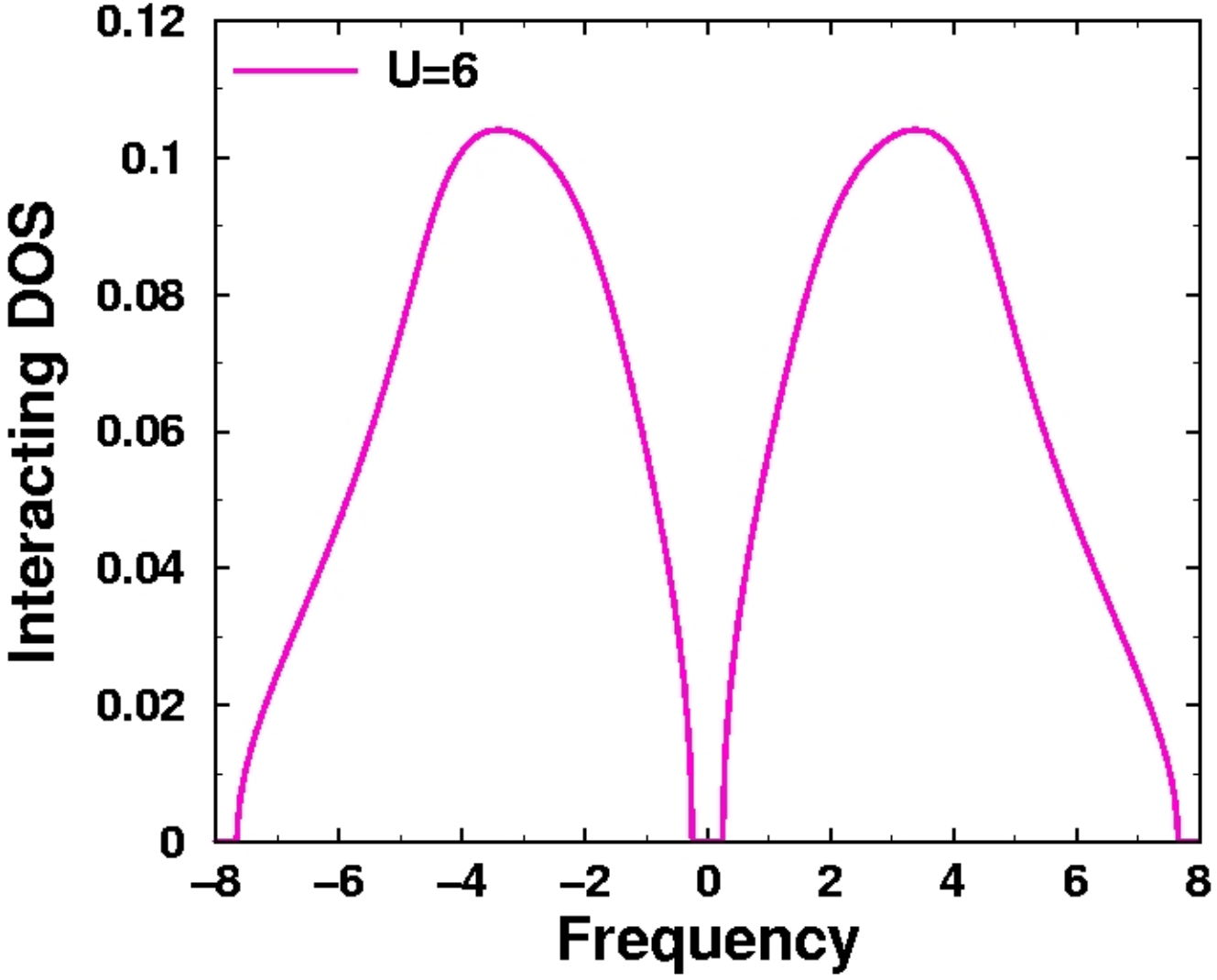
Metal-insulator transition (half-filling)



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.

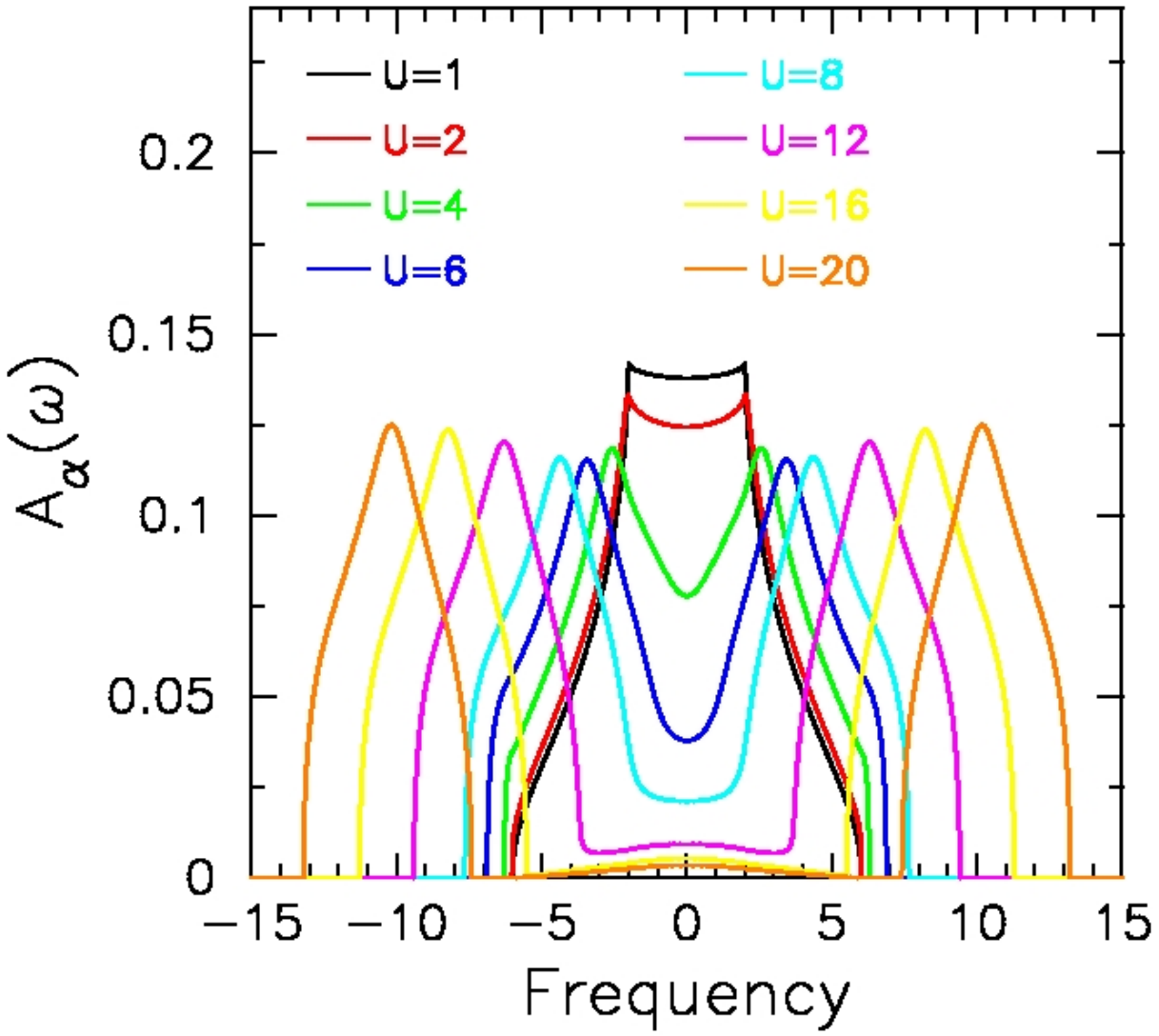
Near the MIT (U=6)



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.

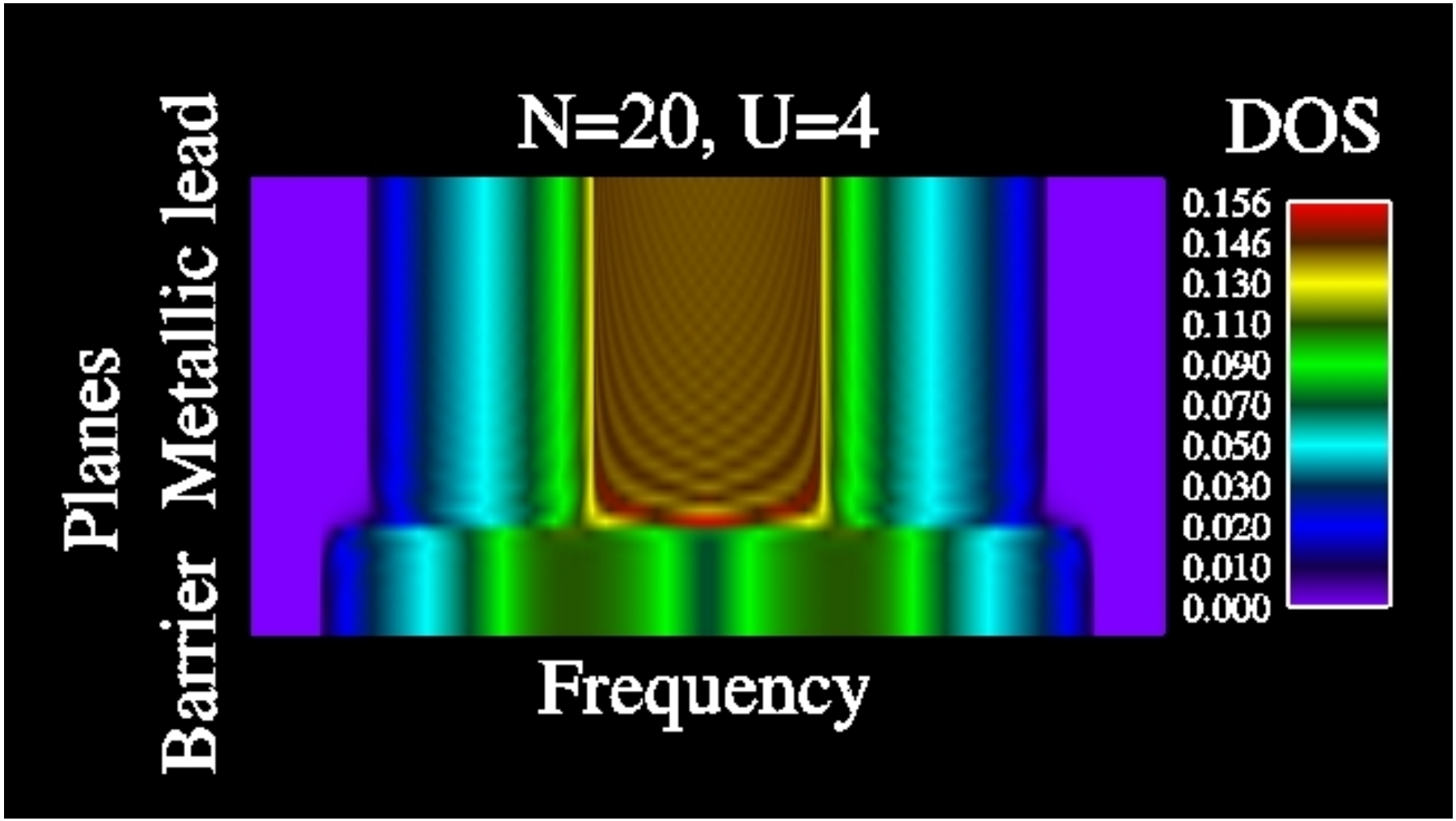
L=a (Single plane barrier)



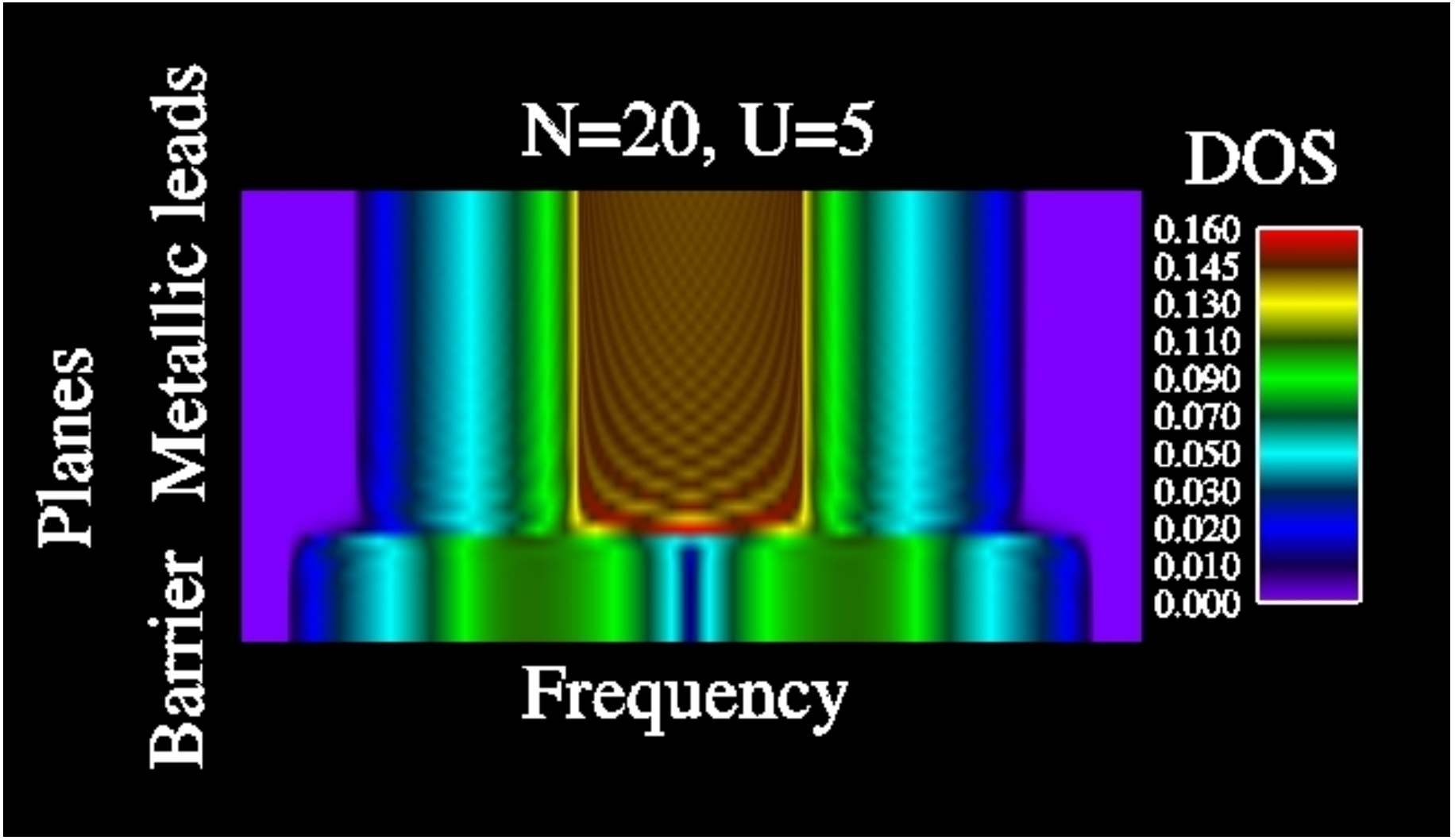
Local DOS on the central barrier plane. Note how the upper and lower Hubbard bands form for the Mott transition, but there is always substantial subgap DOS from the localized barrier states.

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

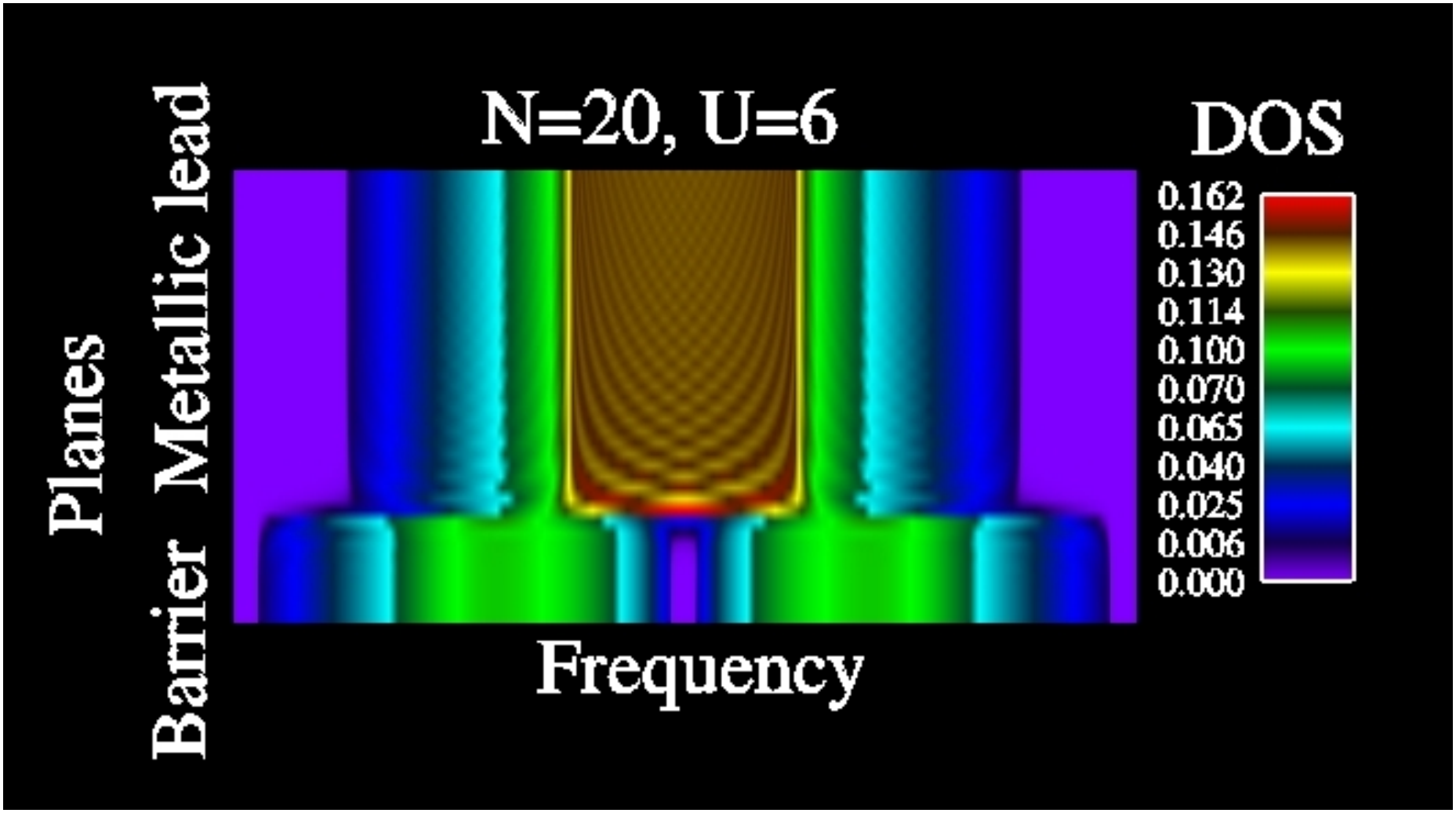
U=4 (anomalous metal) DOS



U=5 (near critical) DOS

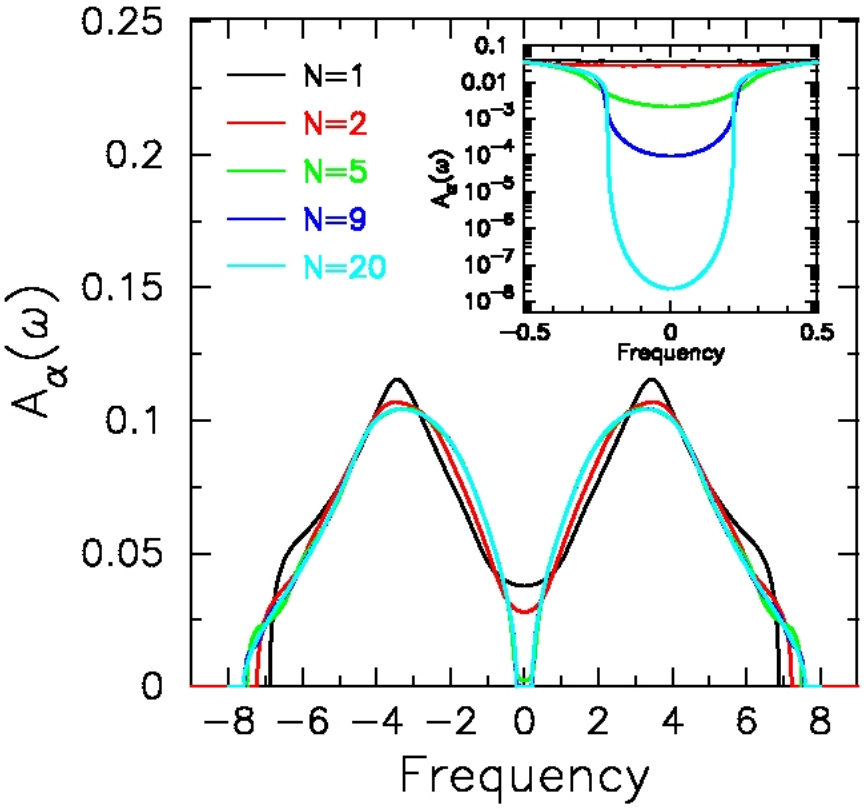


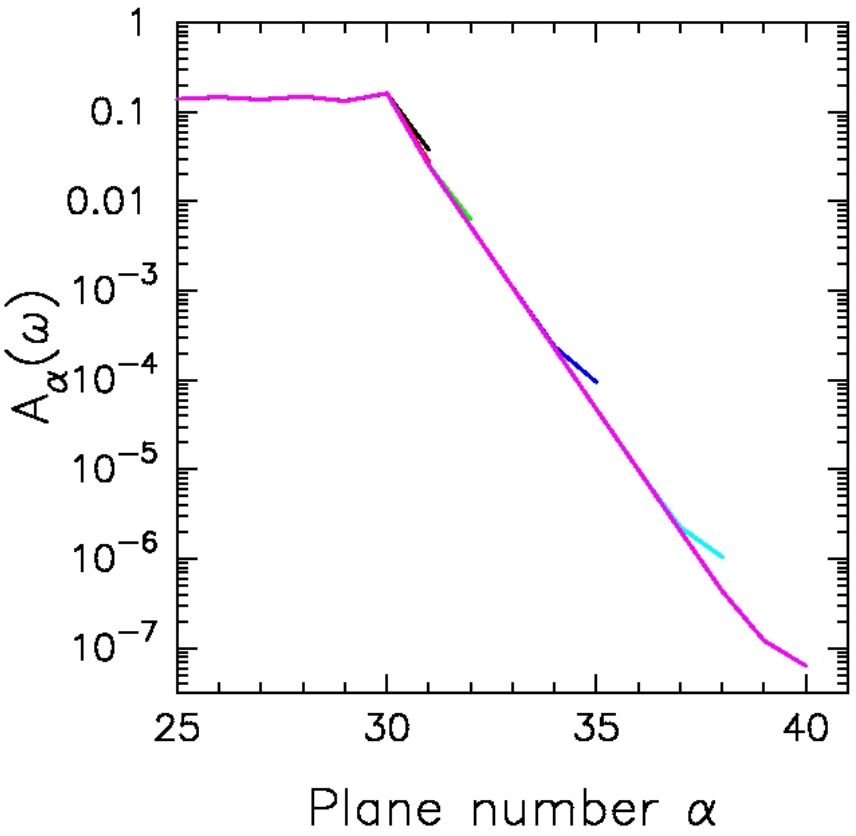
U=6 (small-gap insulator) DOS



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 …

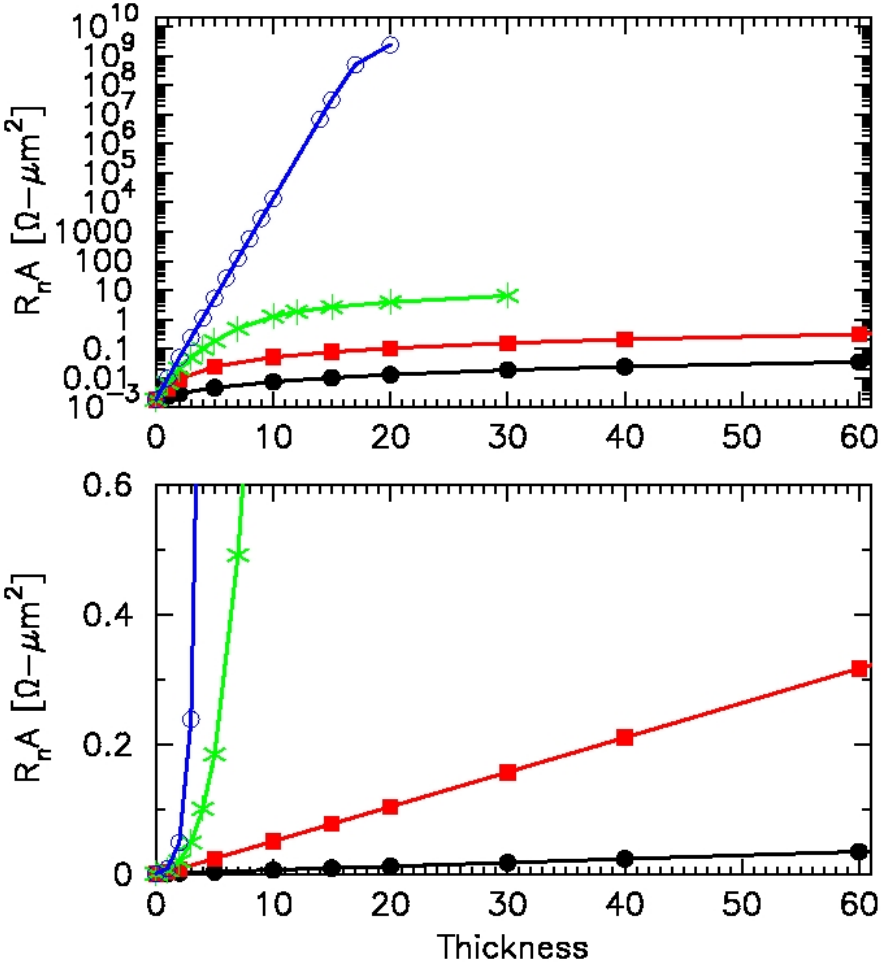
Junction resistance

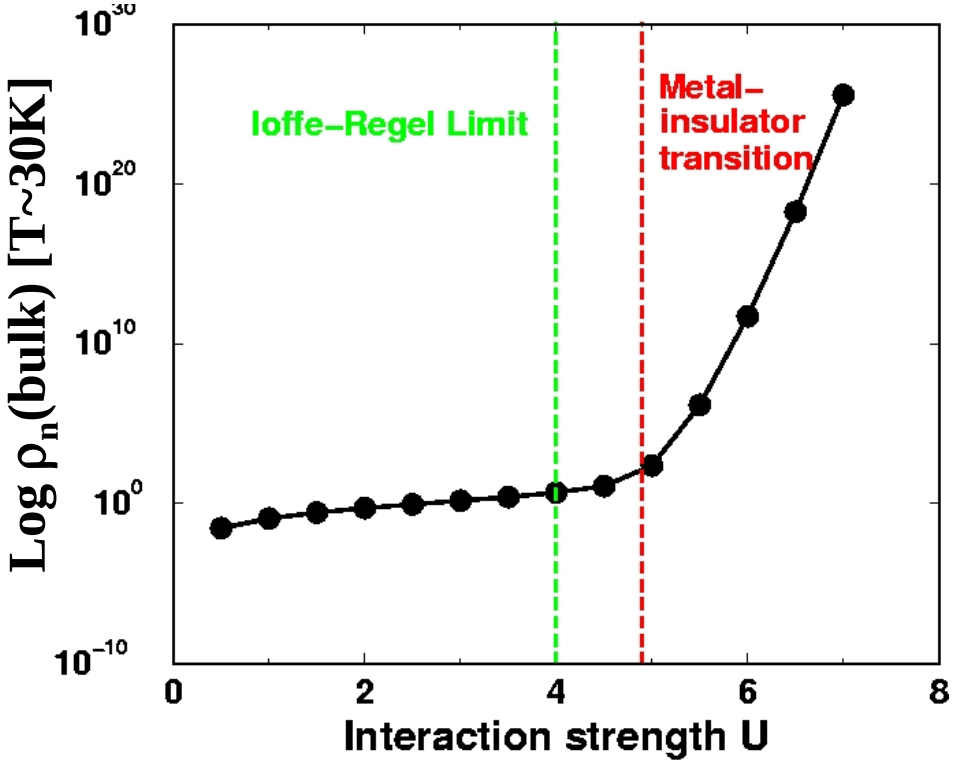
- The linear-response resistance can be calculated in equilibrium using a Kubo-Greenwood approach.
- We must work in real space because there is no translational symmetry.
- R_n is calculated by inverting the conductivity matrix and summing all matrix elements of the inverse.

Junction resistance (derivation)

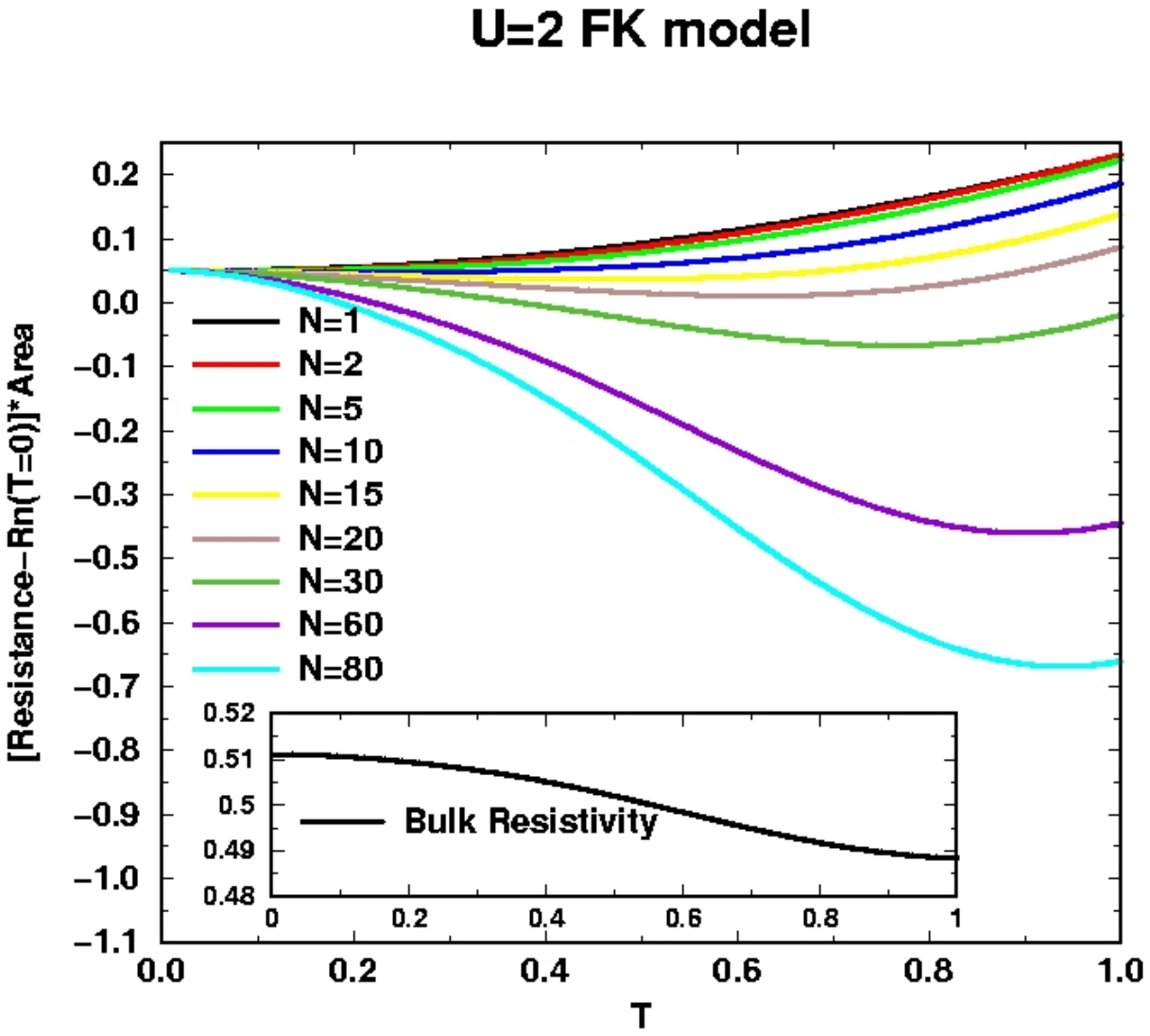
- Maxwell's equation gives $\mathbf{j}_i=\sum_j \sigma_{ij}\mathbf{E}_j$ where the index denotes a plane in the layered device. (The field at plane j causes a current at plane i.)
- Taking the matrix inverse gives $\mathbf{E}_i=\sum_j \sigma^{-1}_{ij}\mathbf{j}_j$; but the current is conserved, so **j** 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





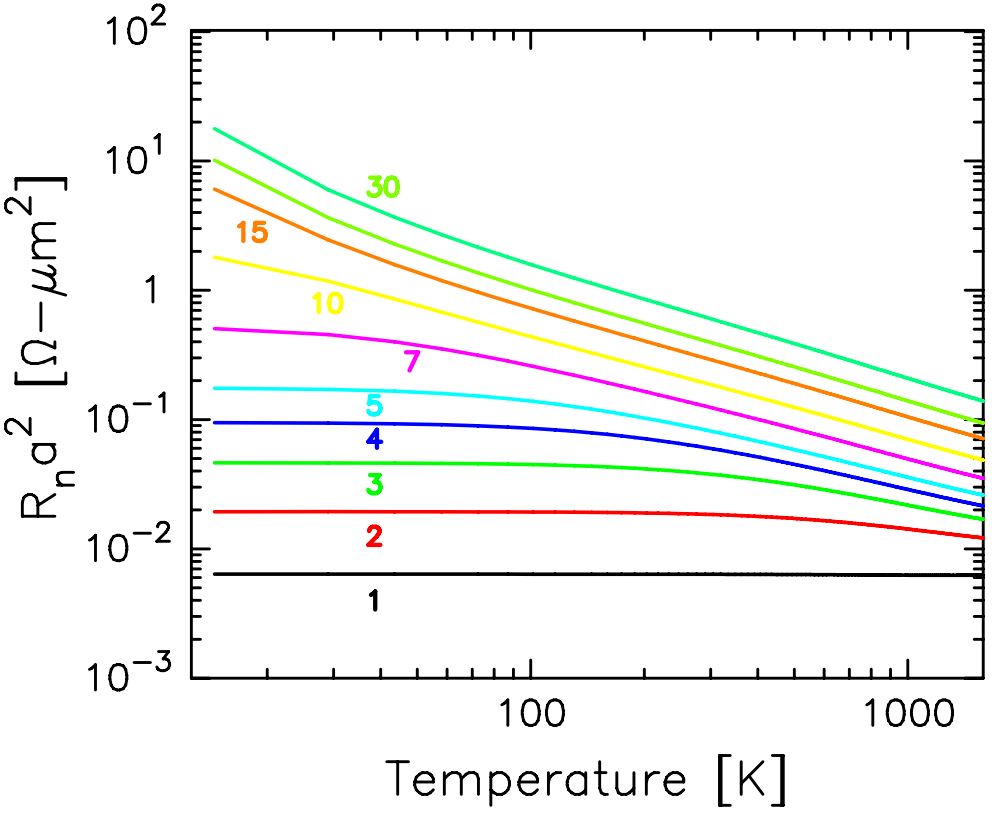
Temperature dependence (correlated metal)



The thin barrier appears more "metallic"; as the barrier is made thicker, the resistance is equal to a contact resistance plus an Ohmic contribution, proportional to the bulk resistivity.

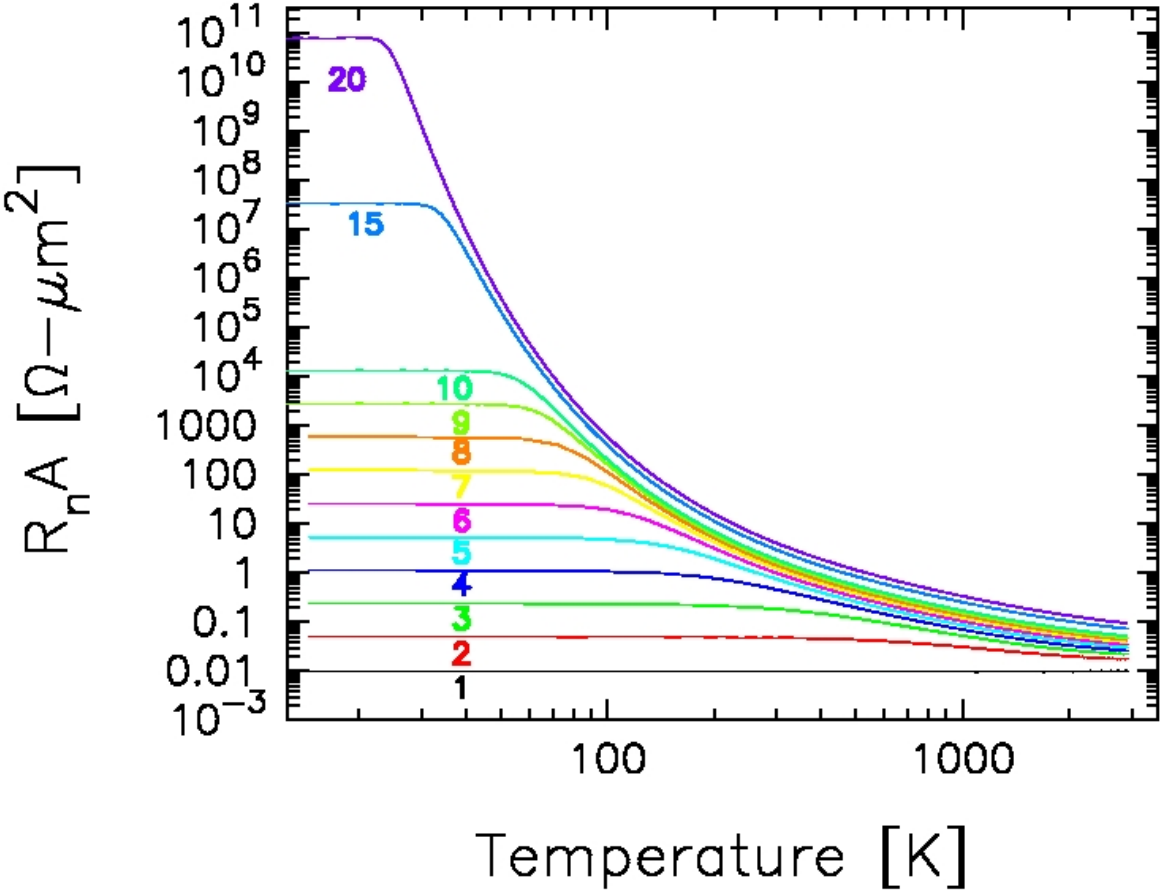
Resistance for U=5 (near critical)

- Tunneling occurs when the junction resistance has little temperature dependence.
- Incoherent transport occurs when the temperature dependence becomes strong.



Resistance for U=6 (correlated insulator)

- Resistance here shows the tunneling plateaus more clearly, and a stronger 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:

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

- 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

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

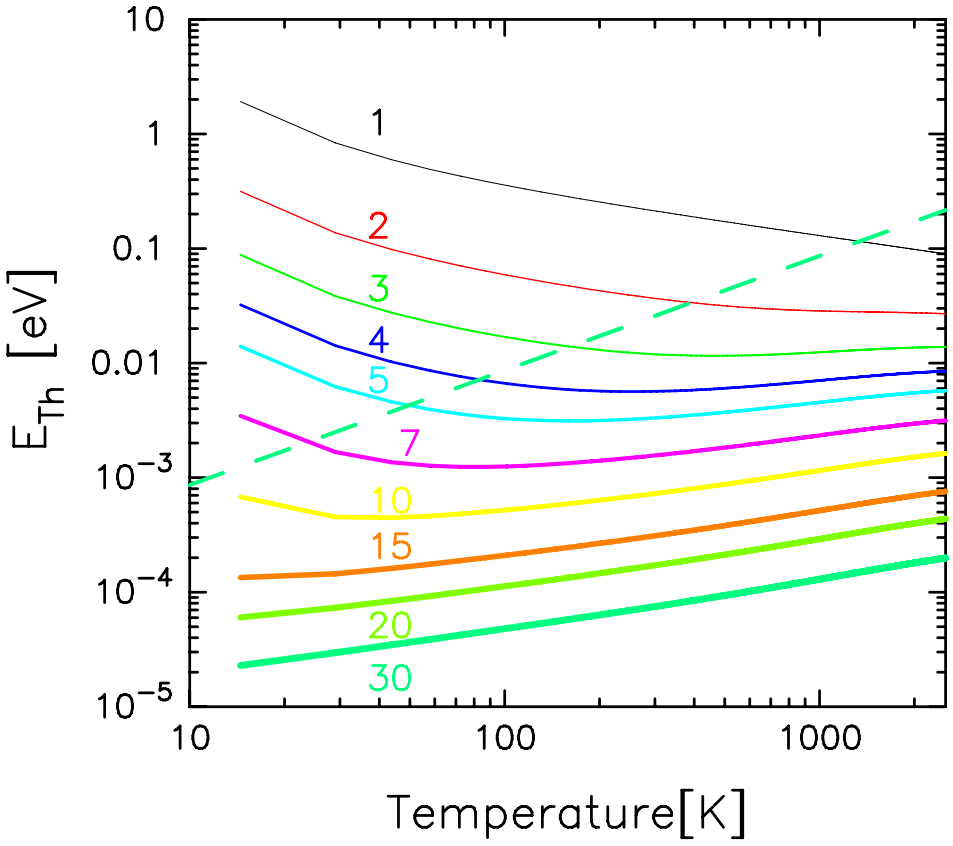
- Generalizing the above relation to an insulator by

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

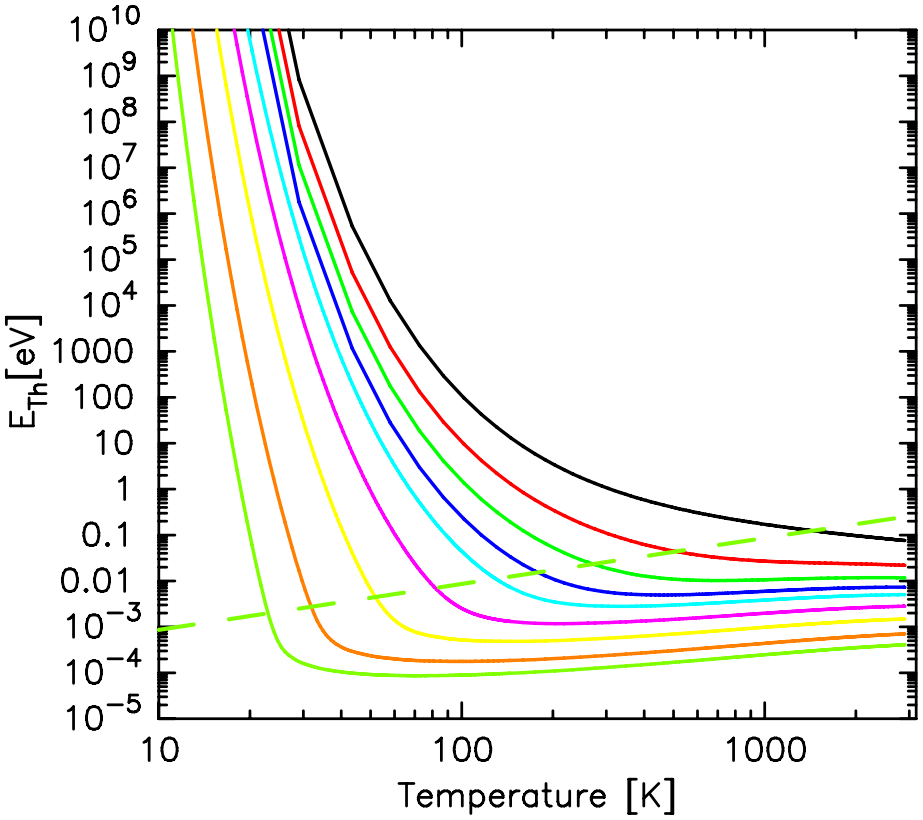
gives the general form for the Thouless energy.

Temperature dependence of E_{Th}

U=5

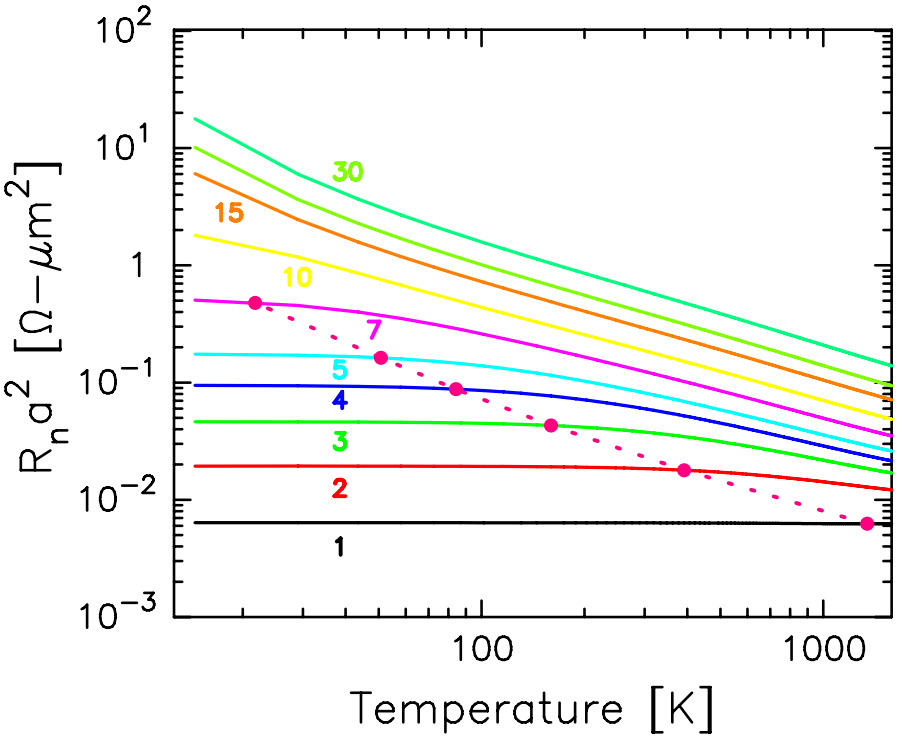


U=6

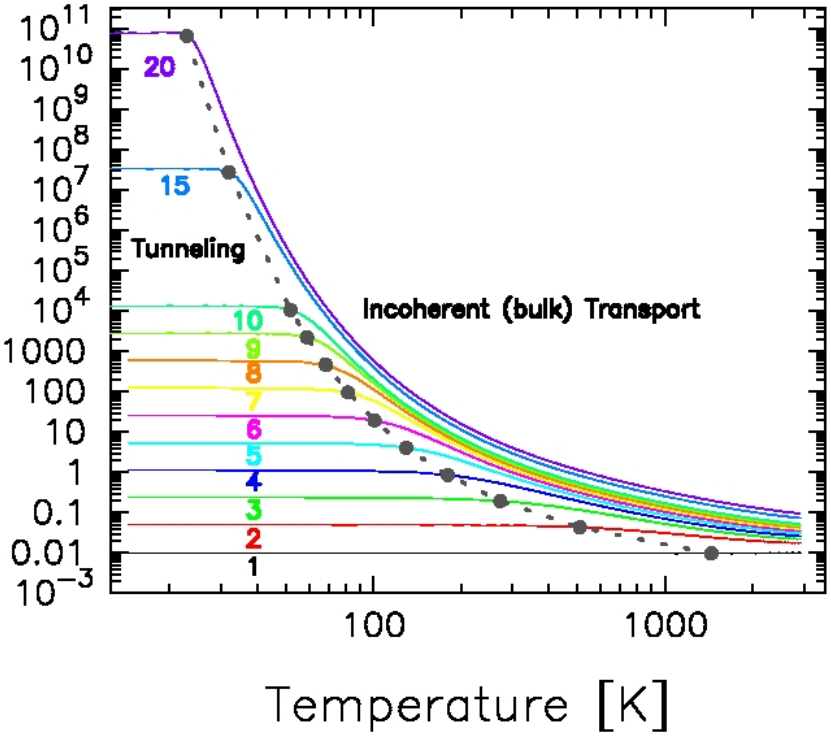


Temperature dependence (II)

U=5 FK model



U=6 FK model

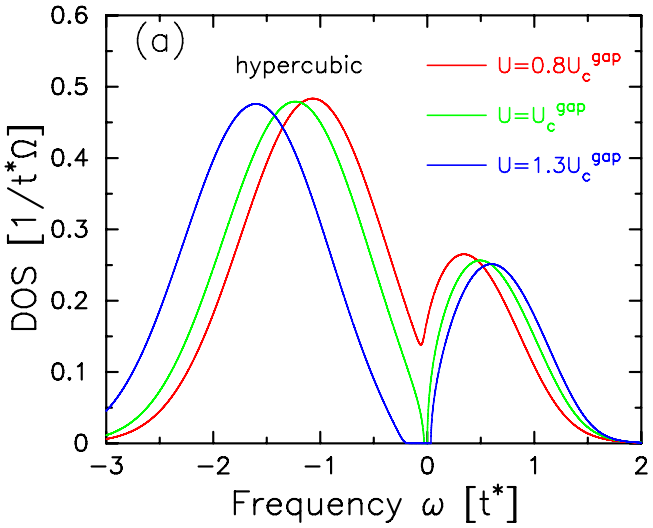


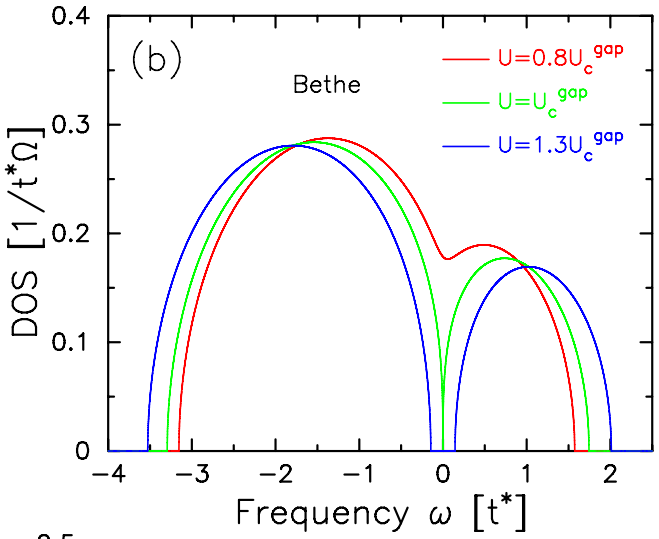
The Thouless energy determines the transition from tunneling to incoherent transport as a function of temperature!

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

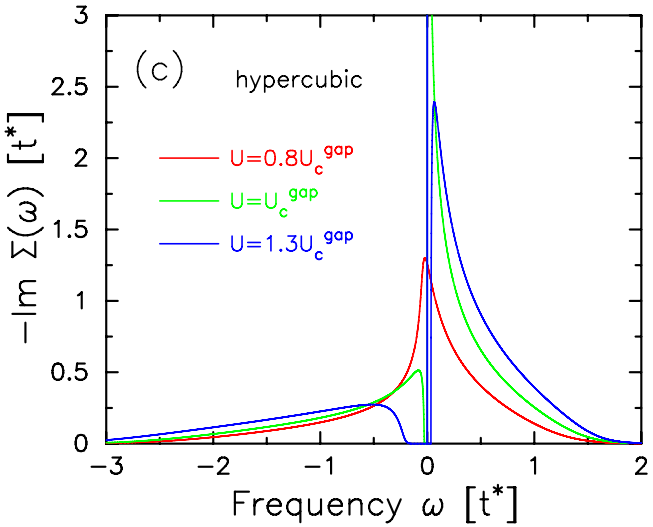
But,
Particle-hole asymmetry is necessary for thermoelectric devices …

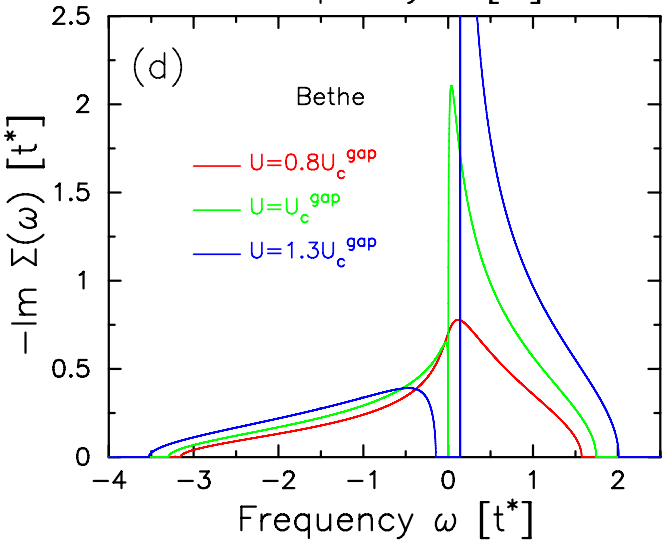
Particle-hole asymmetric MIT





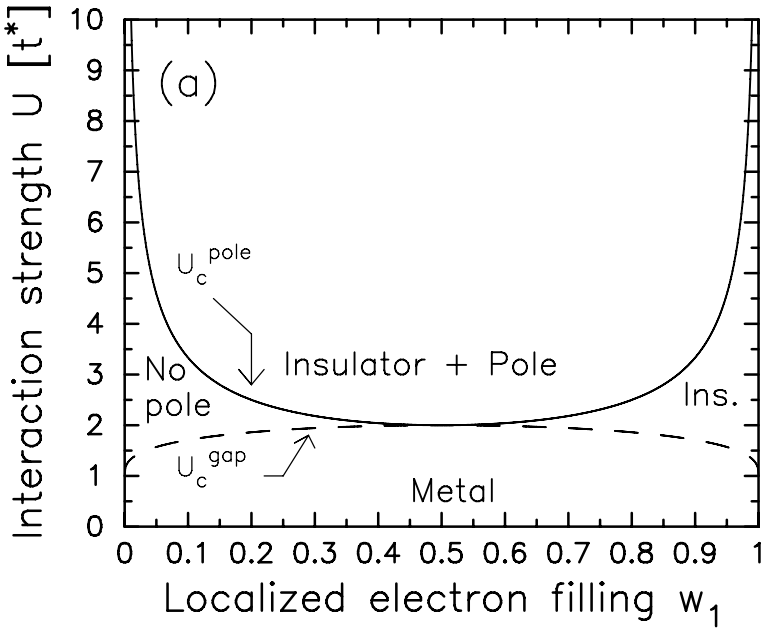
DOS





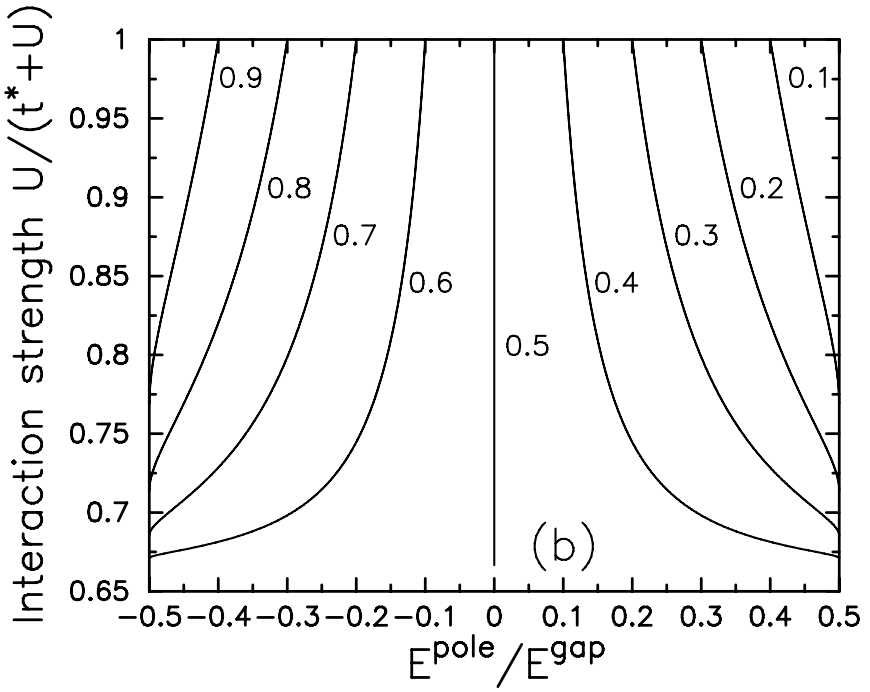
Self-energy

Pole formation and the MIT

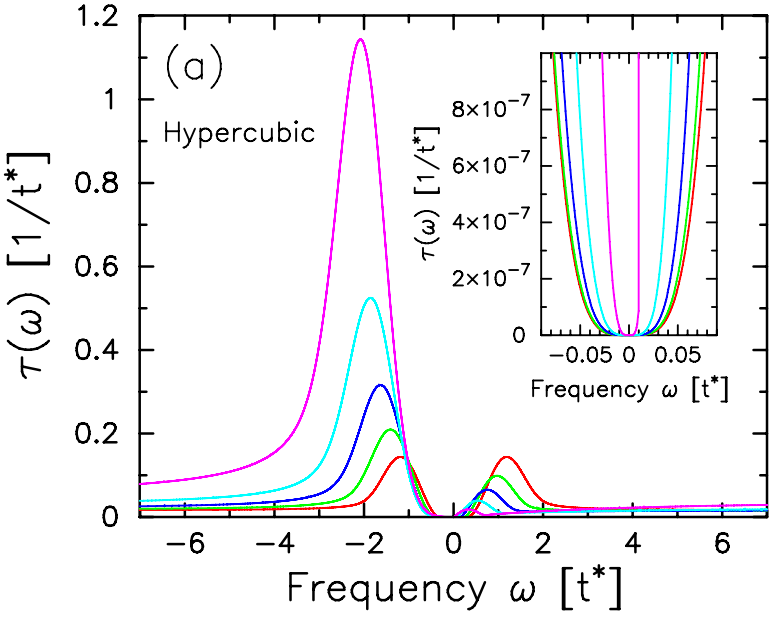


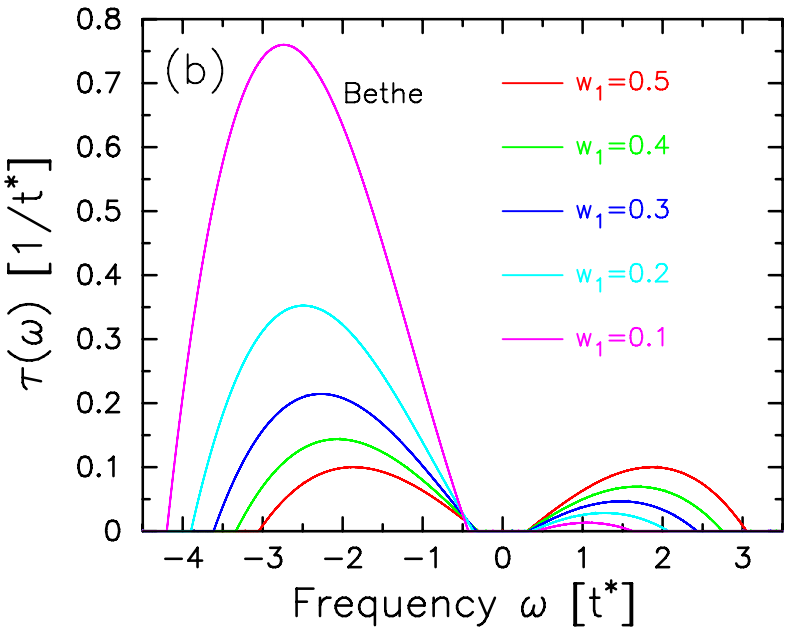
On the hypercubic lattice, the MIT and pole formation in the self-energy coincide. On the Bethe lattice, the pole forms after the MIT except at half filling.

On the Bethe lattice, the pole enters from one band edge, and migrates closer to the center of the gap as U is increased. The pole appears to have no effect on the transport.



Exact relaxation time for transport



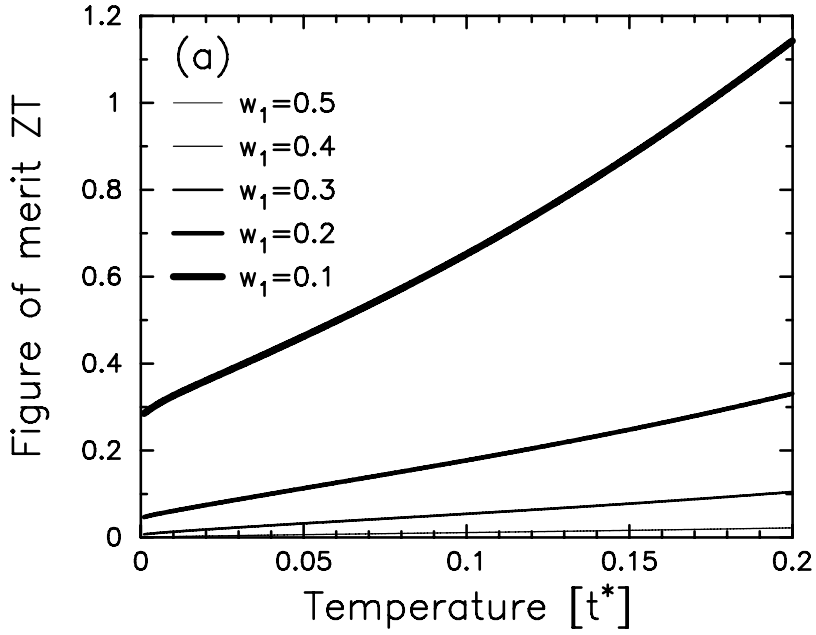


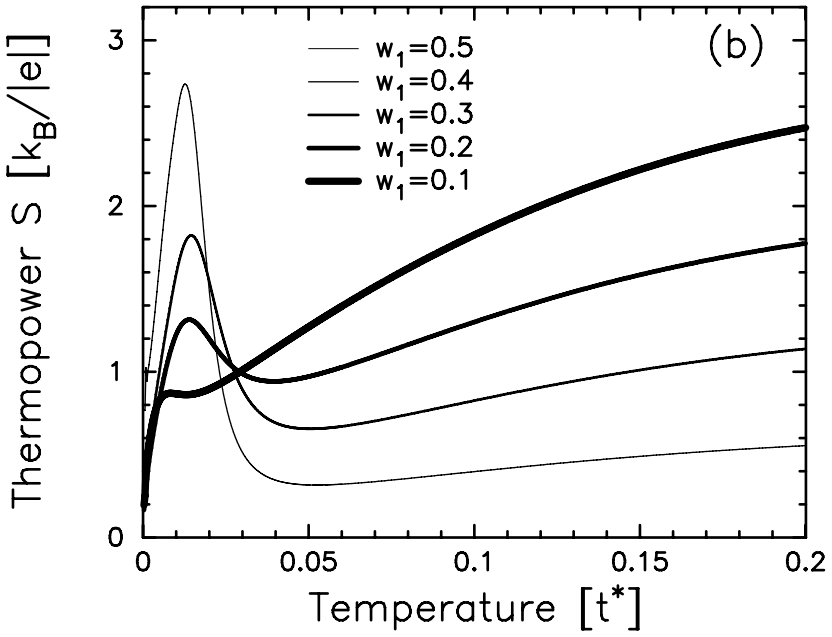
On the hypercubic lattice, the relaxation time has anomalous behavior like quartic dependence in the gap region and a constant value at large frequencies.

On the Bethe lattice, the relaxation time behaves as expected---it vanishes in the gap and it vanishes outside the band.

In both cases, the Jonson-Mahan theorem can be employed to find the thermal transport.

Thermopower and ZT





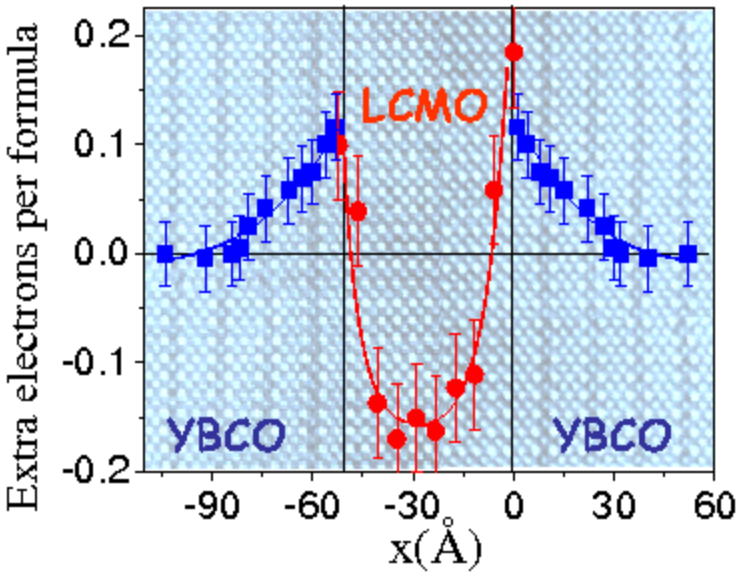
On the Bethe lattice, the figure of merit can be nonzero at T=0 if the system is insulating. It typically grows with T, yielding applications more in the power generation spectrum than for cooling.

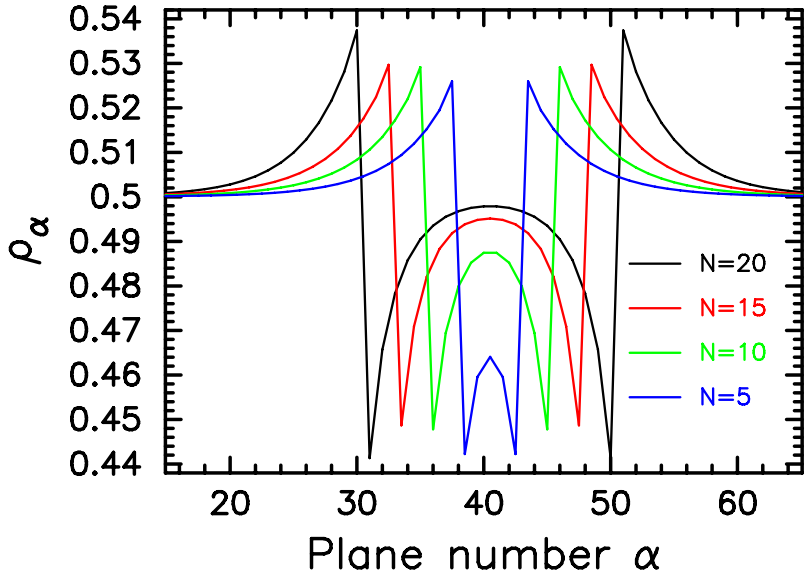
On the Bethe lattice, the thermopower can have a sharp peak at low T for a small-gap insulator close to half filling.

When we have particle-hole asymmetry, we must have an electronic charge reconstruction at the interfaces …

The chemical potential is set by the bulk leads. If the barrier is at a different chemical potential in the bulk, then the device will form screened dipole layers at each interface transfering charge from the metal to the barrier, or vice versa. This is similar to the well-known Schottky barrier in semiconductor devices.

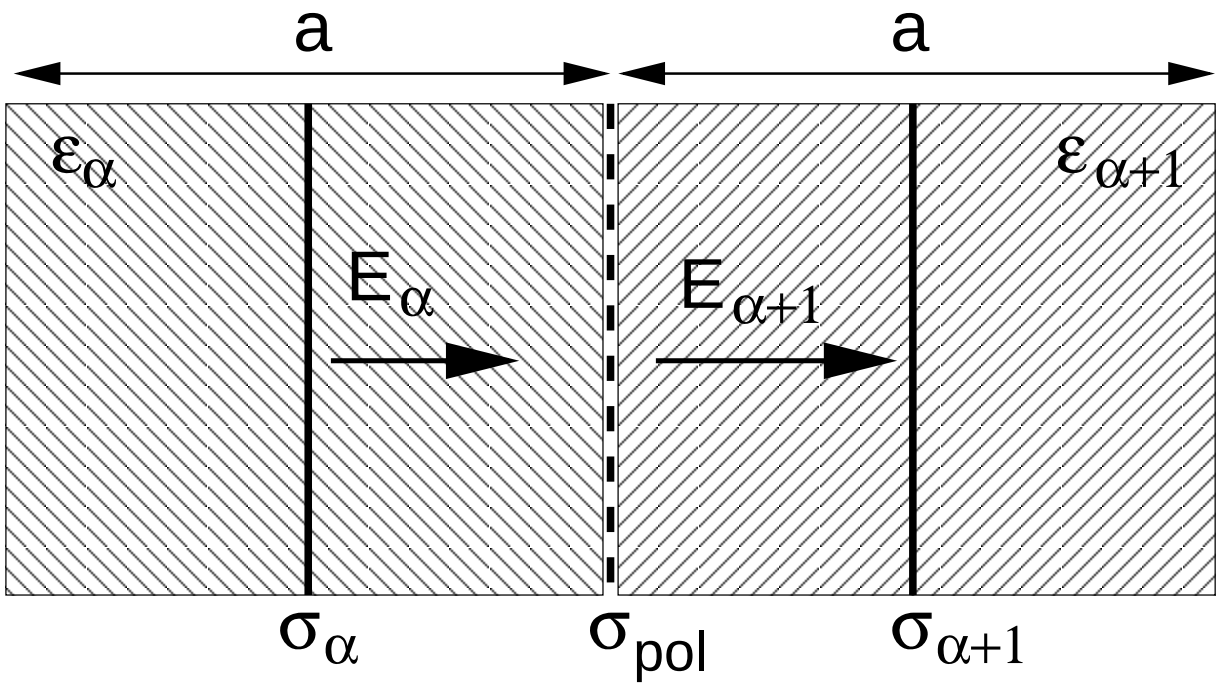
Electronic charge reconstruction





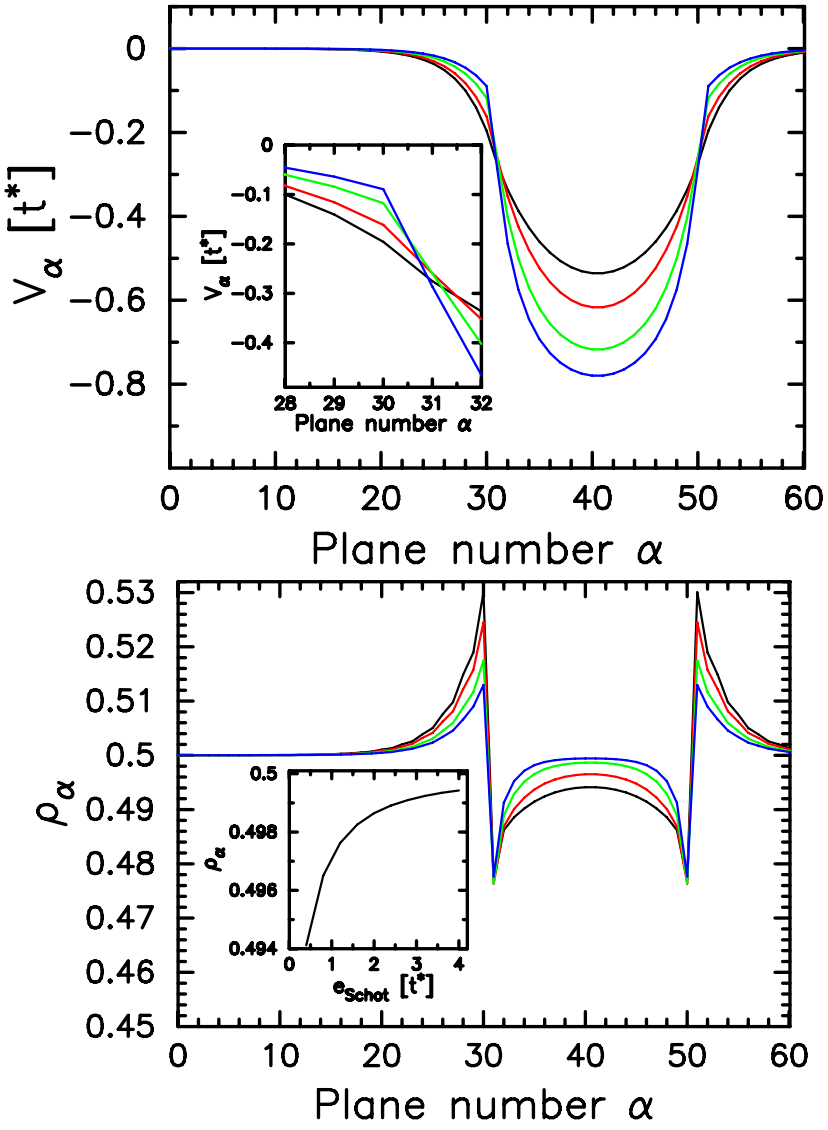
Using a scanning transmission electron microscope with electron energy-loss spectroscopy, one can directly measure the electronic charge at each plane of a strongly correlated multilayered nanostructure. Left are experimental results by Varela et al. on YBCO/LCMO heterostructures, right is a simple theory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

- There are a number of significant challenges to determining the DOS when there is a charge rearrangement.
- The most important is that the integrand for the local Green's function develops poles that must be handled in a principal-value sense. The number and location of these poles varies from plane to plane and from iteration to iteration.
- *We do not yet have numerical results for the DOS in this case.*

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 four important cases: the Seebeck effect, thermal conductivity, and a refrigerator/generator, the Peltier effect.

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 because it is a constant!

Thermal conductance

For a thermal conductance measurement, we also 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 algebra is the same as before, but now we examine the ratio of the heat current carried through the device to the change in the temperature of the device:

Hence, $\Delta T=-\sum_{ij}M^{-1}_{ij}\, j_Q$ and the thermal conductance is

$$\kappa=-j_Q/\Delta T= 1/\sum_{ij}M^{-1}_{ij}\ ;\ M=L^{22} - L^{21}(L^{11})^{-1}L^{12}$$

Note the similarity to the resistance calculation for the charge transport---now we must use the matrix that yields the effective heat transport of the device!

Refrigerator/generator

For a device, we also isolate the device but now drive electrical current through the system. *Hence there is no heat created or destroyed in the steady state (i.e., the heat current is conserved)*:

The algebra is similar to before, but now we must include the charge current j:

We find that ΔT and ΔV are linear functions of j and j_Q, with complicated coefficients that are functions of the matrices L^{11}, L^{12}, L^{21}, L^{22}, and M. The figure of merit can be written as

$$ZT = TS^2/R_n\kappa = T\,(\Delta V/\Delta T)^2/(\Delta V/j)/(j_Q/\Delta T)$$
$$= T\,(\Delta V/\Delta T)\,(j/j_Q).$$

Peltier effect

The Peltier effect is quite different from the cases examined so far. In this case, the temperature of the device is kept fixed by contact with a thermal bath (like immersion in a liquid refrigerant). Charge current flows through the device, and the heat current varies from plane to plane. The total change in the heat current through the device yields the amount of heat that is exchanged with the reservoir to maintain the constant temperature profile.

The algebra is quite simple now---since the charge current is conserved, we find the heat current satisfies

$$j_{Qi} = \sum_{jk} L^{21}_{ij} (L^{11})^{-1}_{jk} j$$

The total change of the heat current is its value on the right minus its value on the left. Dividing J_{QR}-j_{QL} by j yields the Peltier coefficient for the nanostructure.

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) electronic charge reconstruction, and how to self-consistently determine the screened dipole layers that lead to Schottky-like barriers; and (iv) the formalism for thermal transport (with results in the bulk).

In the future, we will complete the charge transport analysis and calculate self-consistent results for important quantities needed for real devices.

This formalism can be generalized to describe systems governed by the Hubbard or periodic Anderson model, and that work is currently underway.