

Thermoelectric response of strongly correlated multilayers

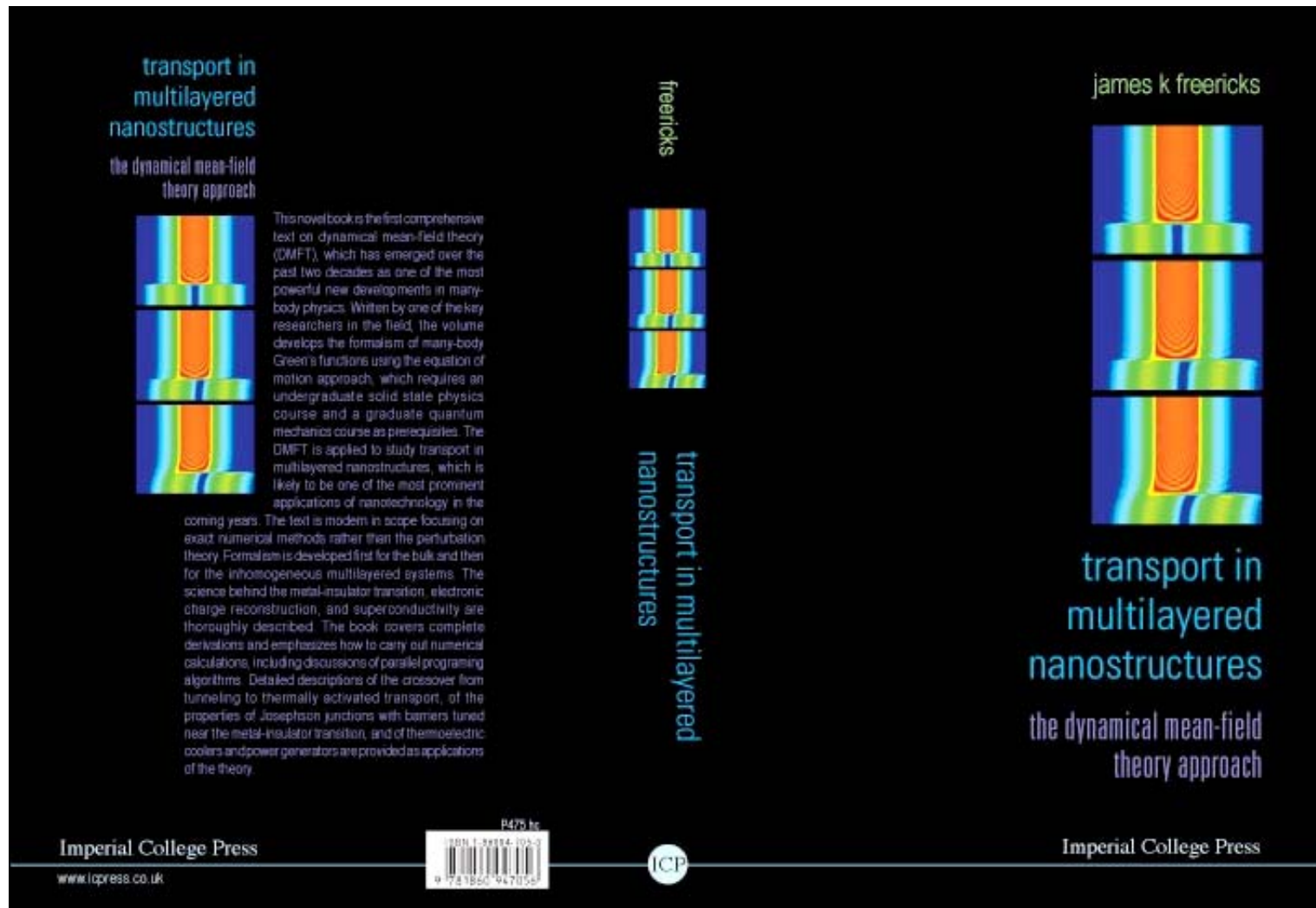
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Funded by the National Science Foundation

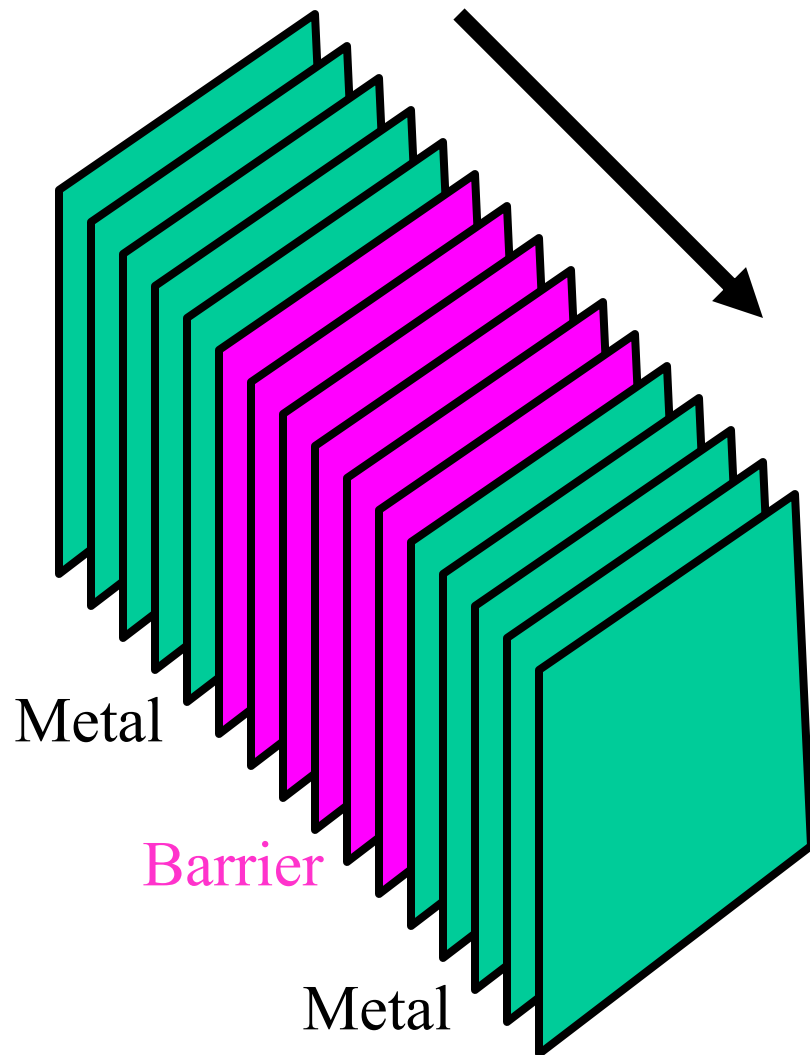
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Graduate-level text published by Imperial College Press



J. K. Freericks, Georgetown University, Hvar Nato Workshop talk, 2008

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 and other oxides.
- Near MIT: V_2O_3 , Ta_xN

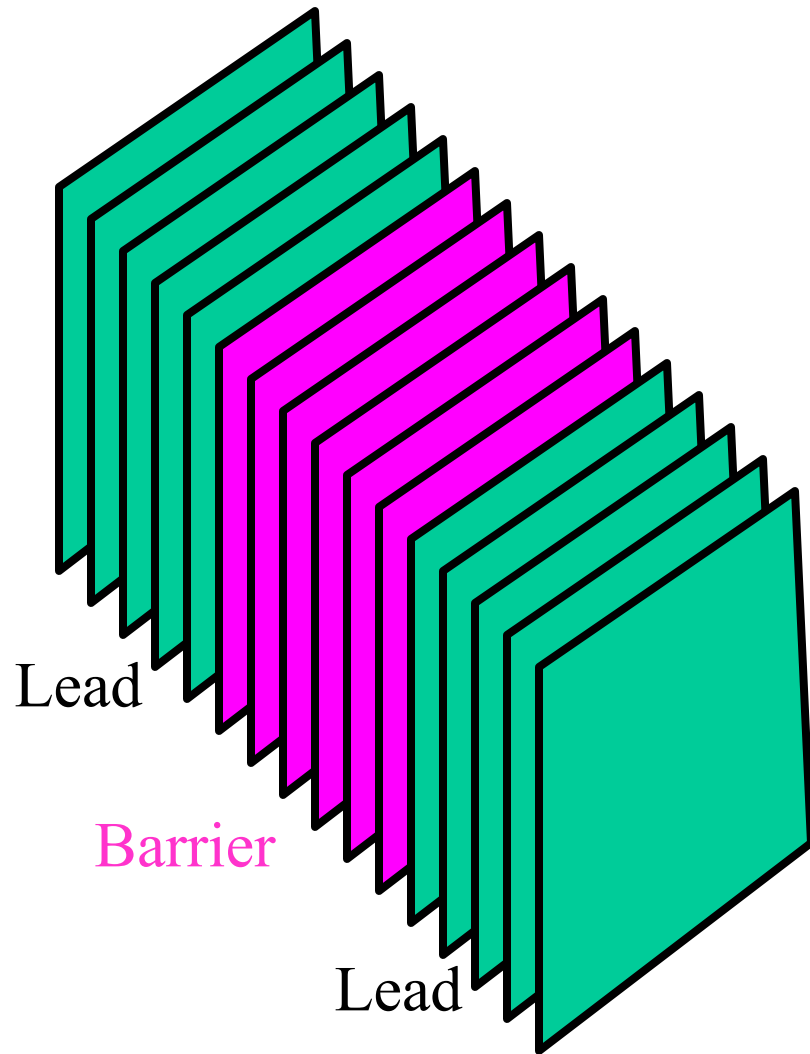
Theoretical Approaches (charge transport)

- 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 ($R_n = h/2e^2 \times [1-T]/T$)
- 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



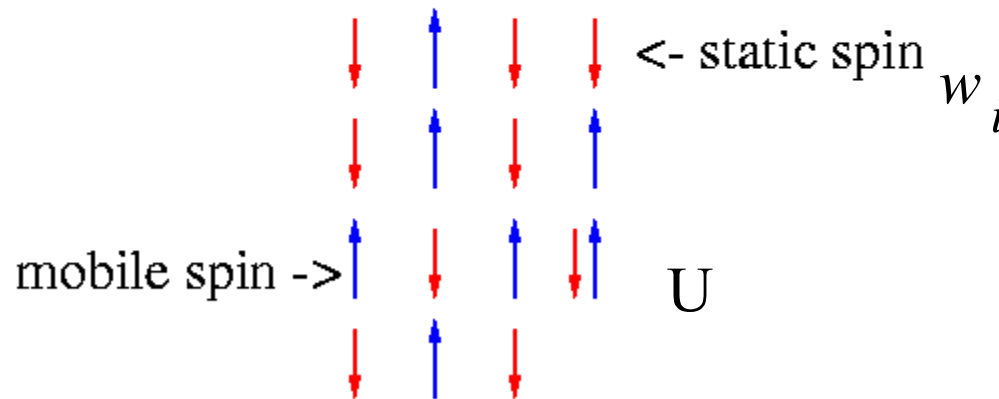
The metallic leads can be ballistic normal metals, mean-field theory ferromagnets, or BCS superconductors.

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

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

Spinless Falicov-Kimball Model and Hubbard Model

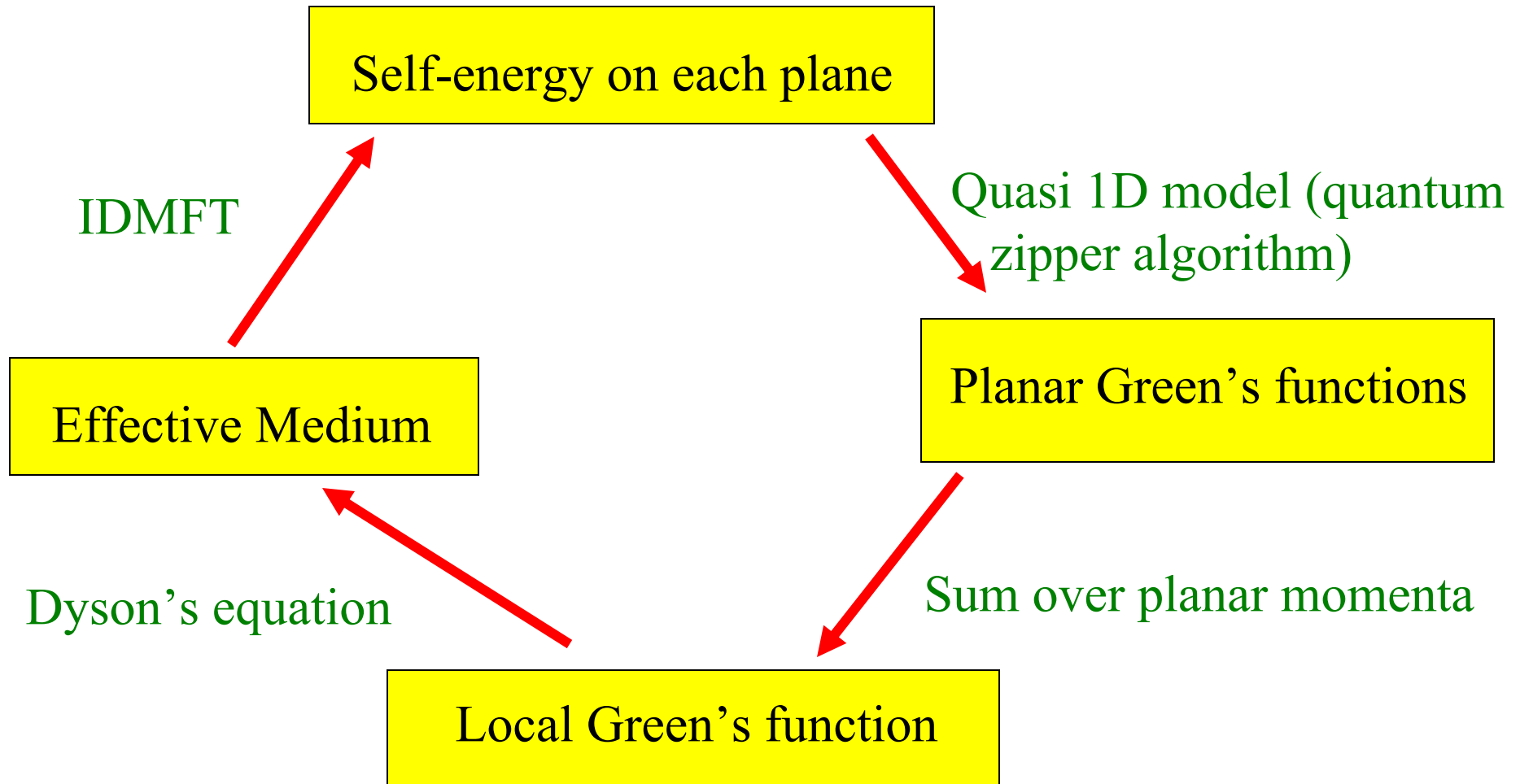
$$H = -\frac{t}{2\sqrt{d}} \sum_{\langle i,j \rangle} c_i^\dagger c_j + E \sum_i w_i + U \sum_i c_i^\dagger c_i w_i$$



For the Hubbard model, the red spins also move.

- **exactly solvable model** in the local approximation using dynamical mean field theory.
- possesses homogeneous, commensurate/incommensurate CDW and SDW phases, phase segregation, and **metal-insulator transitions**.
- *A self-consistent recursive Green's function approach solves the inhomogeneous many-body problem (Potthoff-Nolting algorithm).*

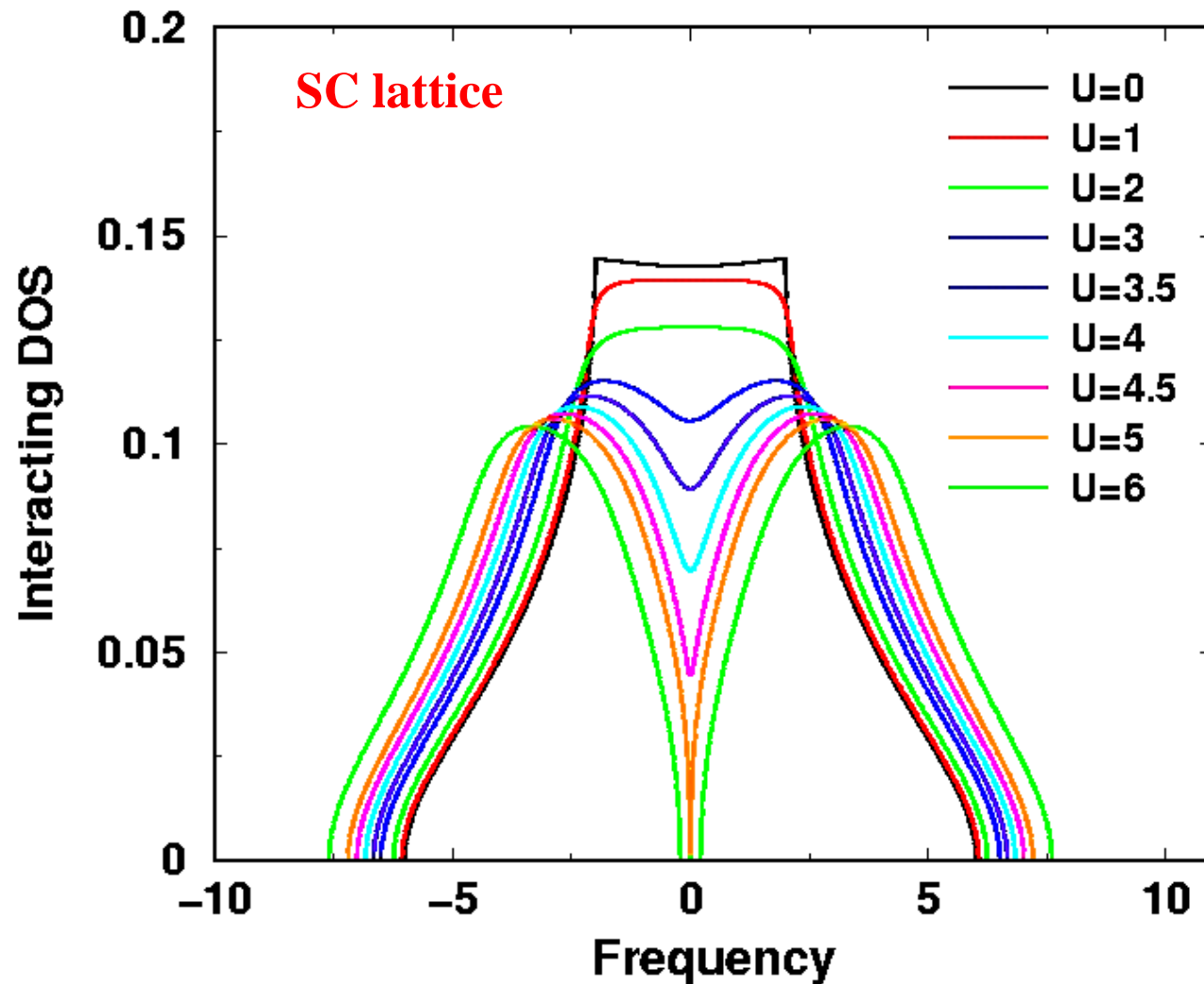
Computational Algorithm



Algorithm is iterated until a self-consistent solution is achieved

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

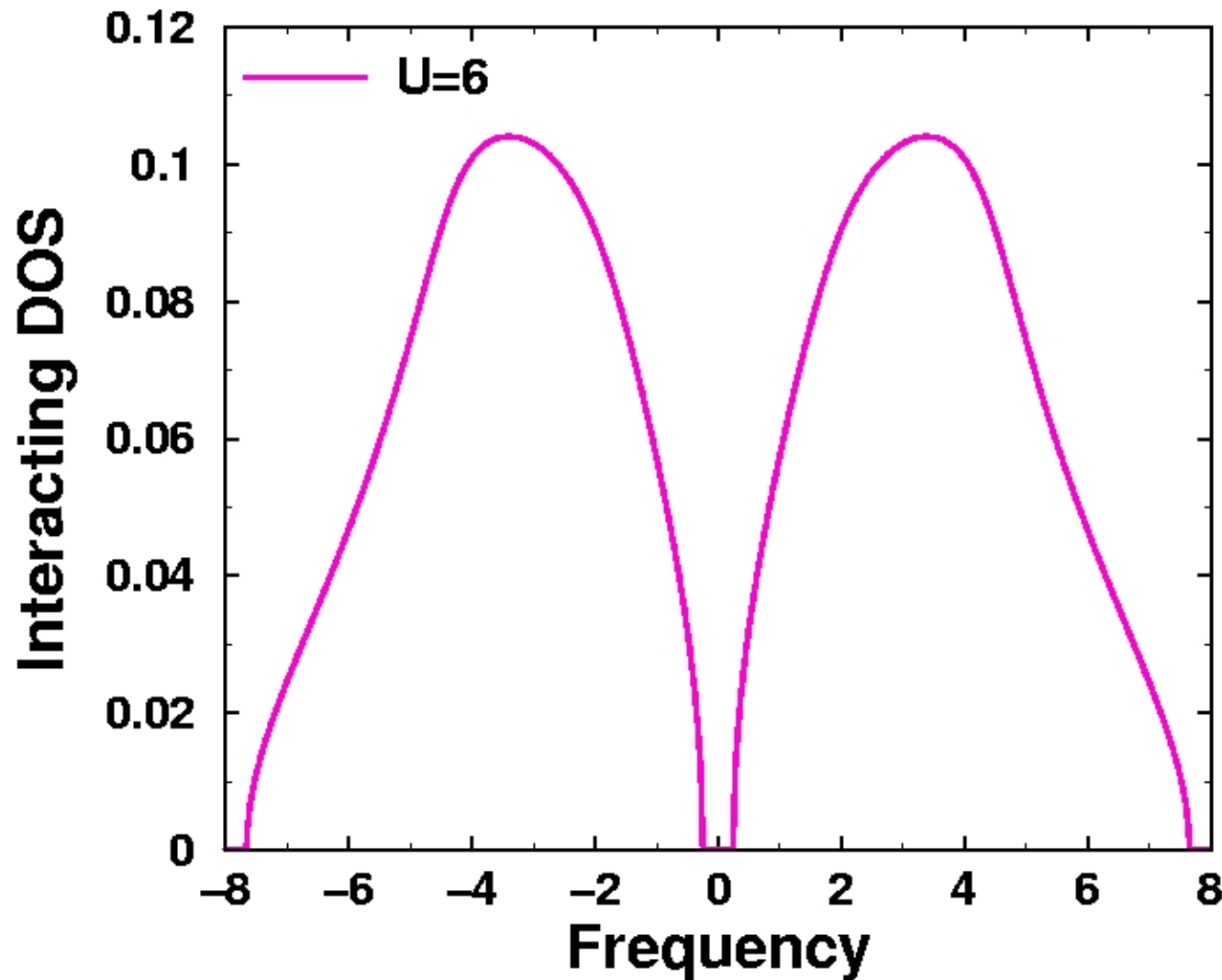
Metal-insulator transition (half-filling)



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 $U=2\sqrt{6}\approx 4.9$ (in the bulk).

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

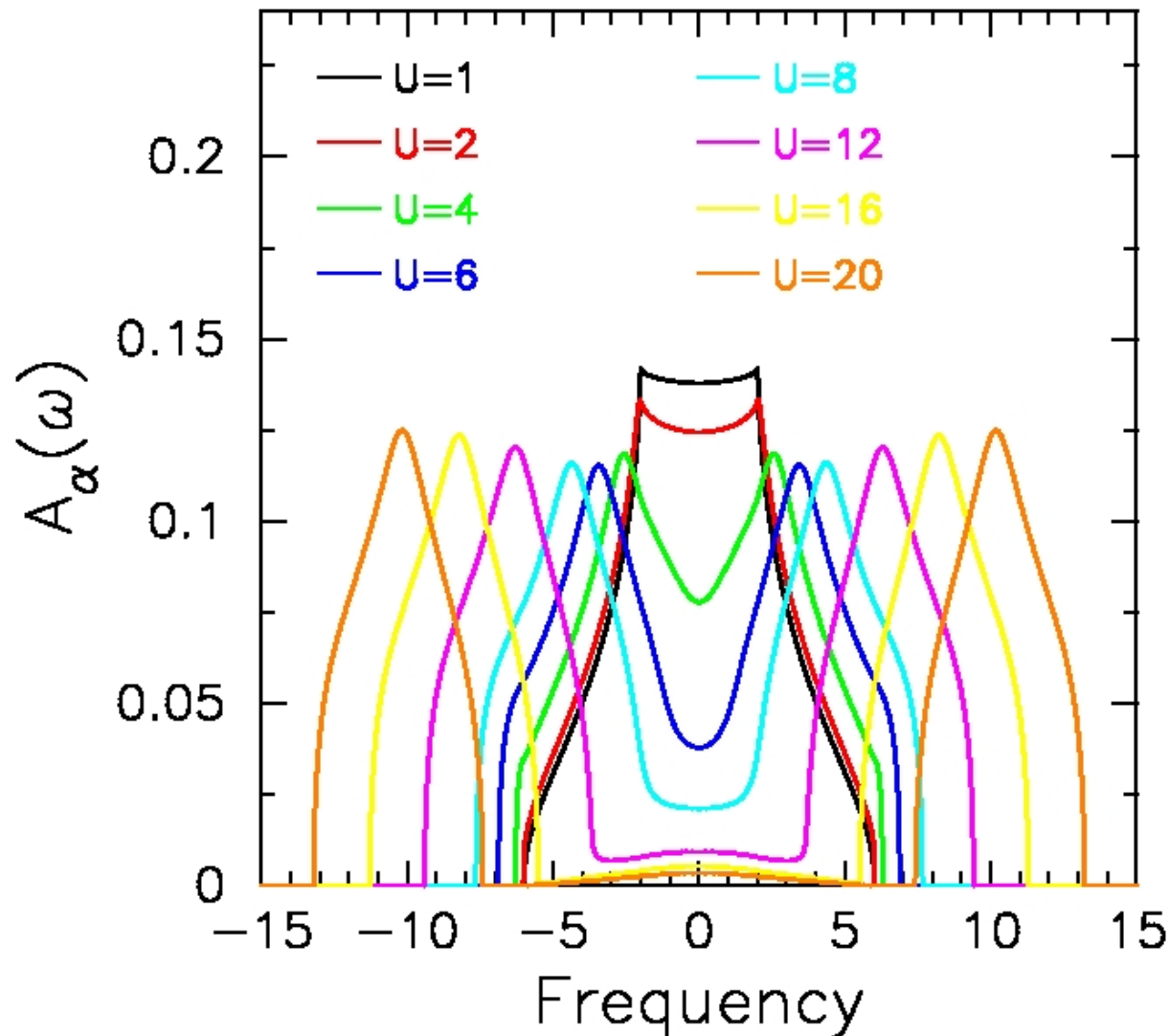
Near the MIT ($U=6$)



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

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

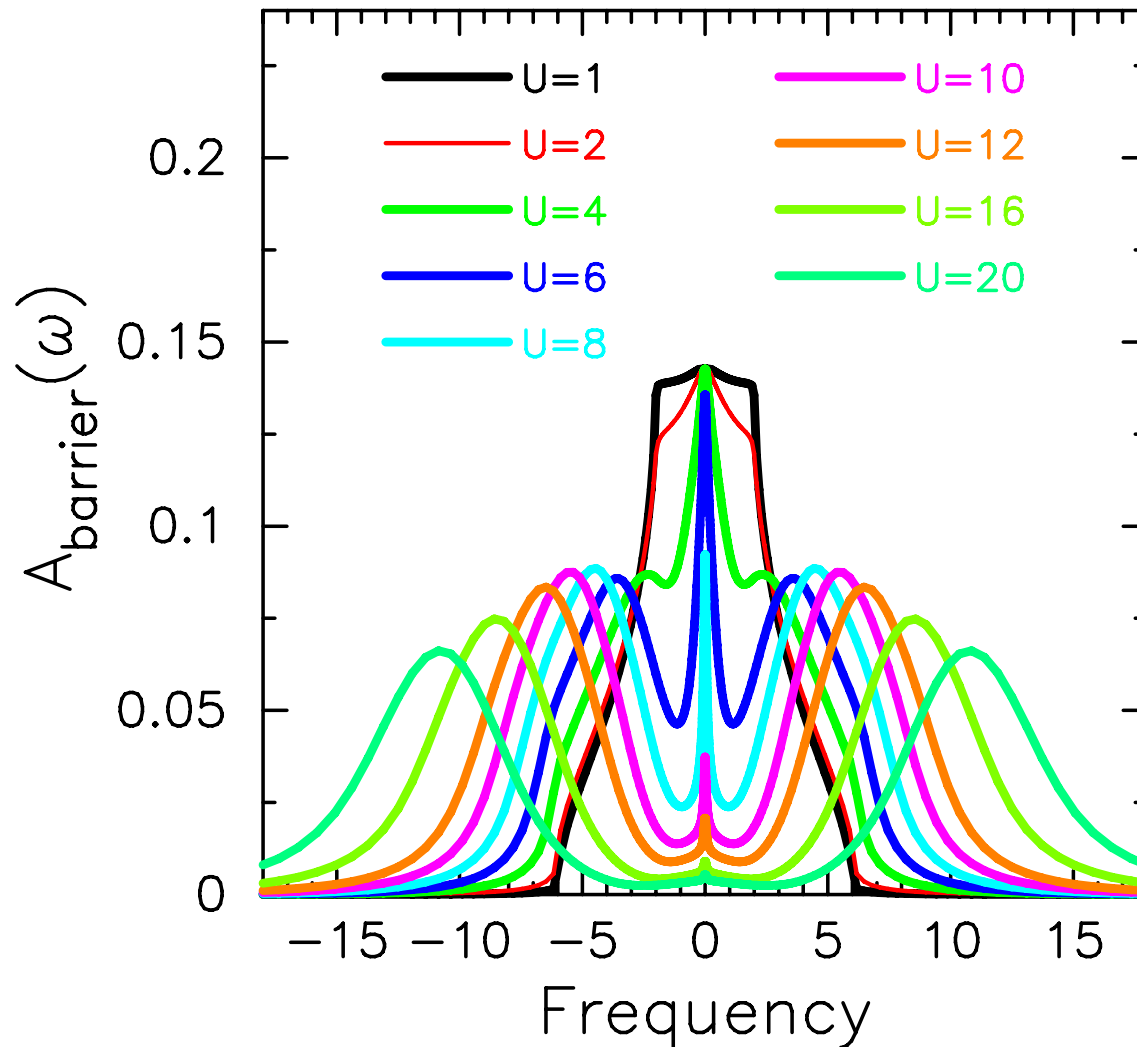
$L=a$ (Single plane barrier, FK model)



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.

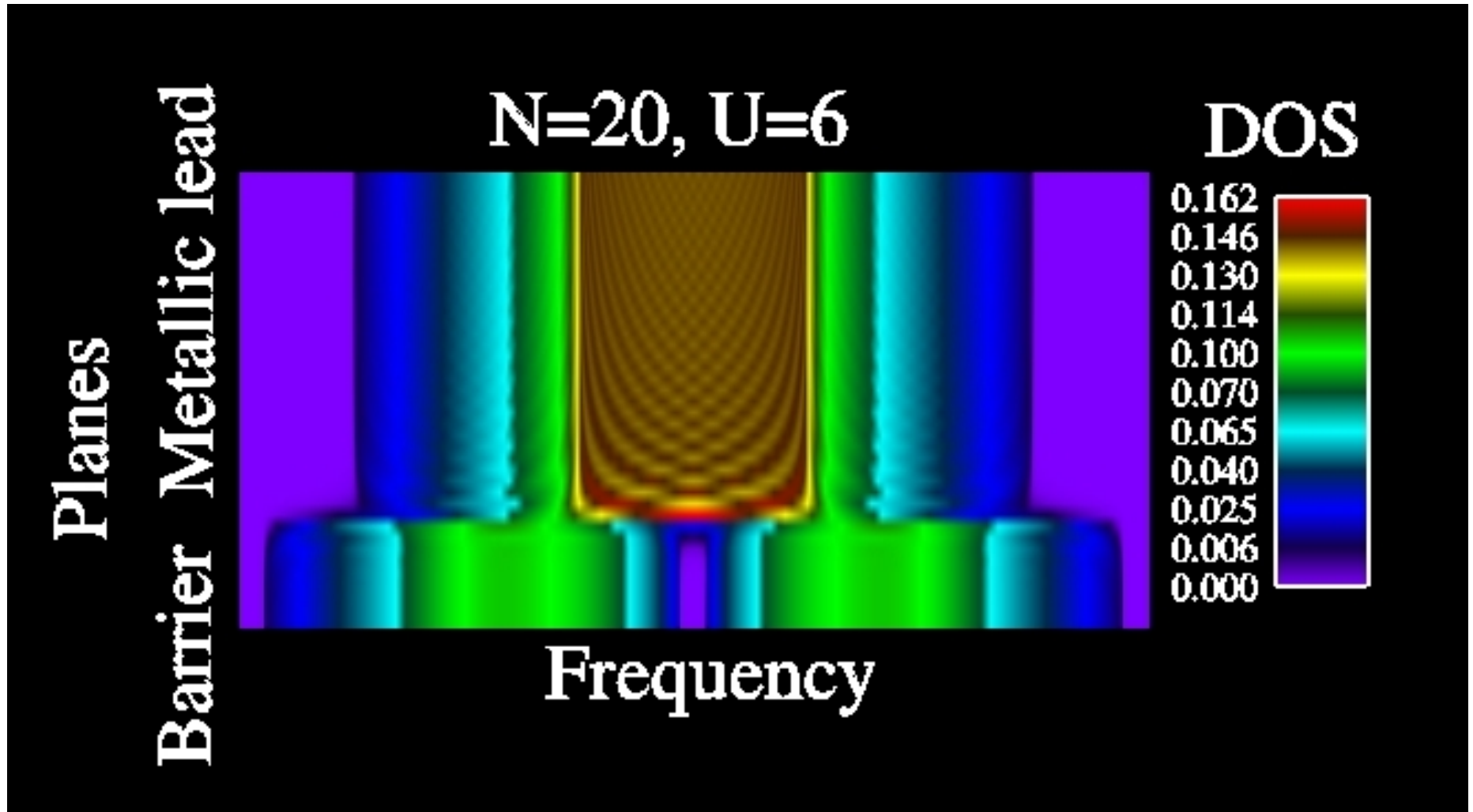
This DOS arises from quantum-mechanical tunneling and has a *metallic shape (i.e. a peak at the center)*.

$L=a$ (Single plane barrier, Hubbard model)

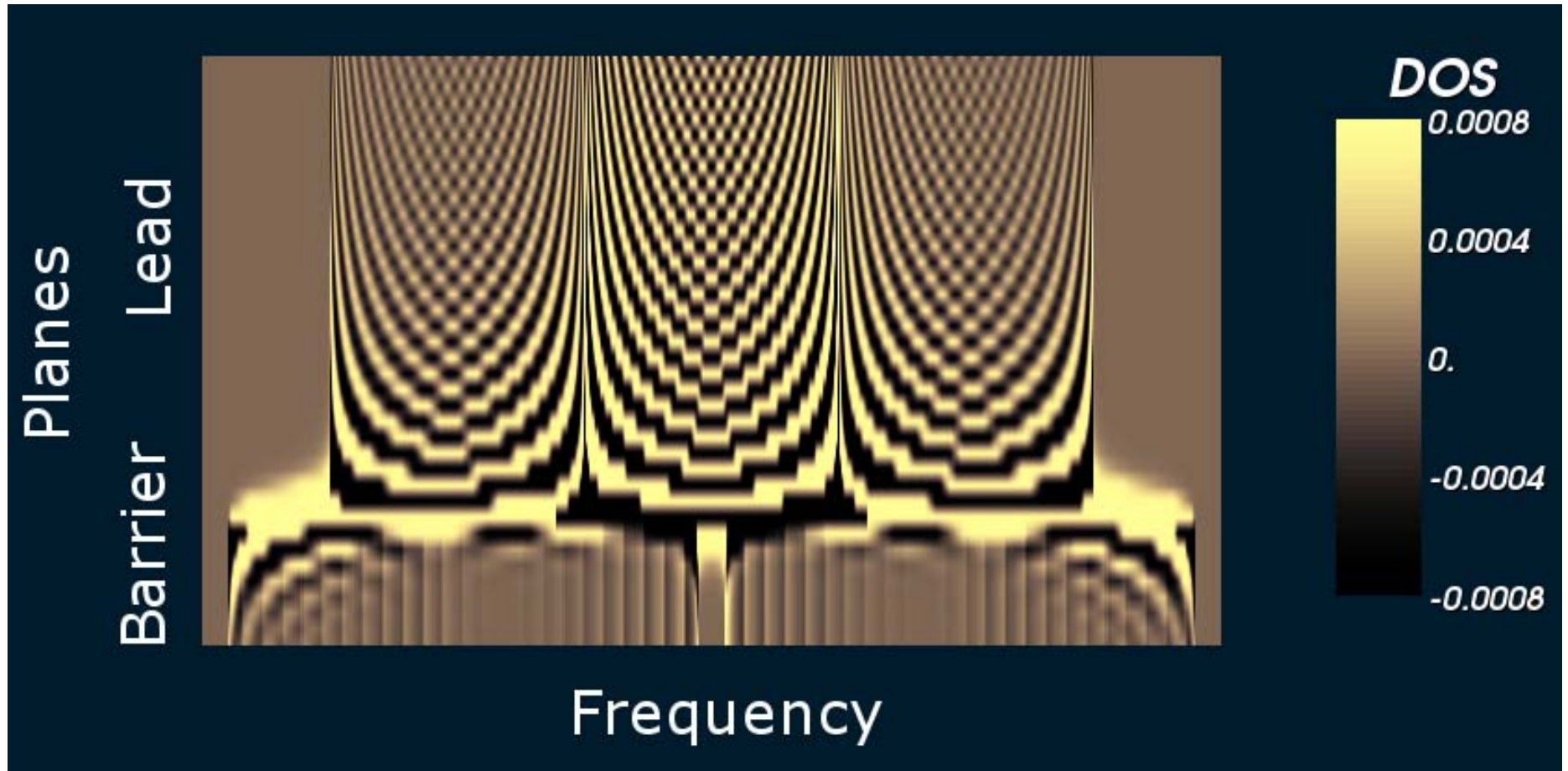


Local DOS on the central barrier plane. Note the similarity with the results for the FK model, except for the coherence peak forming at the lowest energies. FL theory says the coherence peak will be present for all U values at $T=0$ (these calculations are for room temperature with $T=0.01$). **We will return to this point later.**

$U=6$ (small-gap insulator) DOS

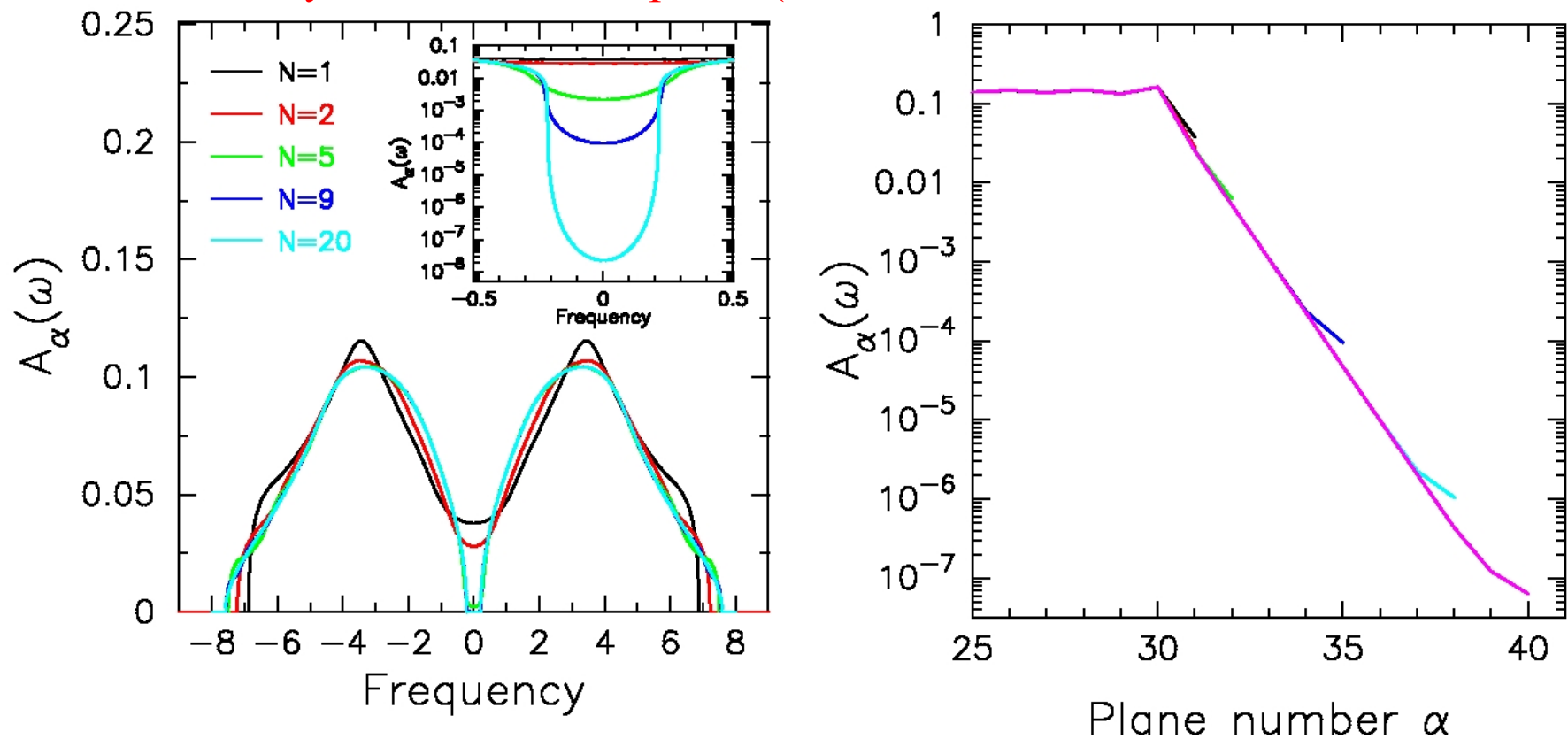


Friedel oscillations induced by interface



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 and is equal to $1/\ln[\text{residue}(U)]$ relating to the residue of the pole in the self-energy of the bulk system in the Mott phase (it is $0.62a$ for $U=6$).



Charge transport and the generalized Thouless energy ...

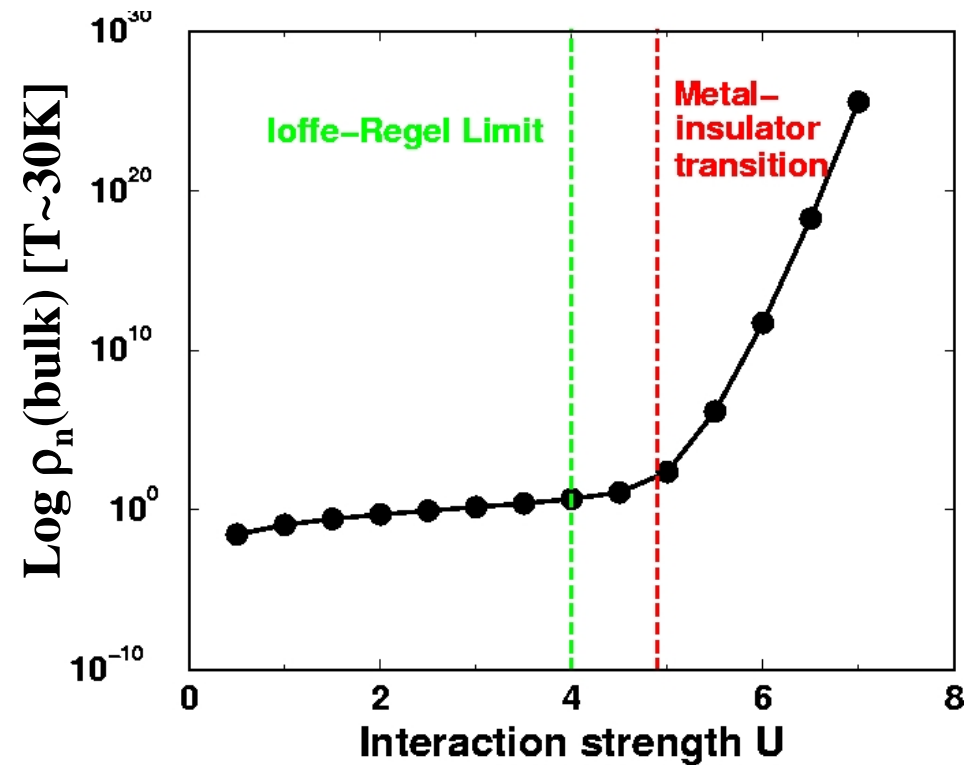
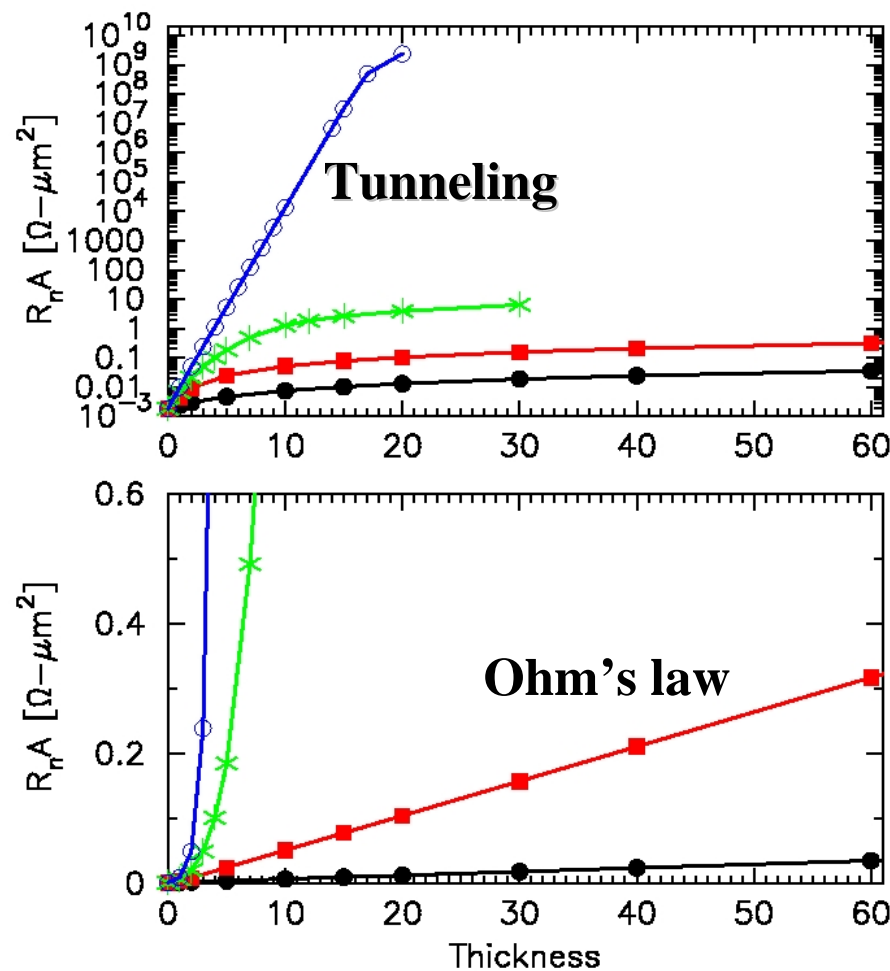
Junction resistance

- The linear-response resistance can be calculated in equilibrium using a Kubo-Greenwood approach (here we neglect vertex corrections).
- 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)

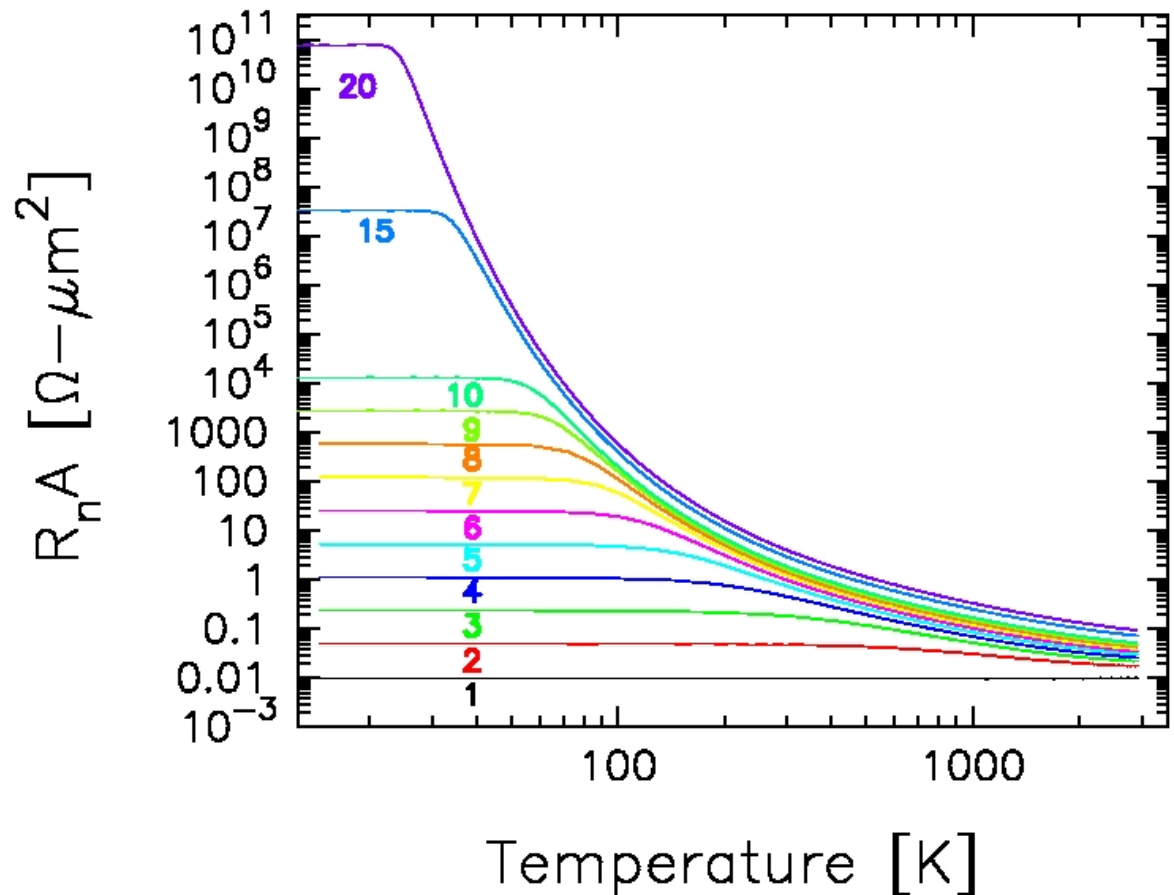
- Maxwell's equation gives $\mathbf{j}_\alpha = \sum_\beta \sigma_{\alpha\beta} \mathbf{E}_\beta$ where the index denotes a plane in the layered device.
(The field at plane β causes a current at plane α .)
- Taking the matrix inverse gives $\mathbf{E}_\alpha = \sum_\beta \sigma^{-1}_{\alpha\beta} \mathbf{j}_\beta$; but the current is conserved, so \mathbf{j} does not depend on the planar index β .
- Calculating the voltage gives $V = a \sum_\alpha E_\alpha = a \sum_{\alpha\beta} \sigma^{-1}_{\alpha\beta} j$, so the resistance-area product is $R_n A = a \sum_{\alpha\beta} \sigma^{-1}_{\alpha\beta}$

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:

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

- 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

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

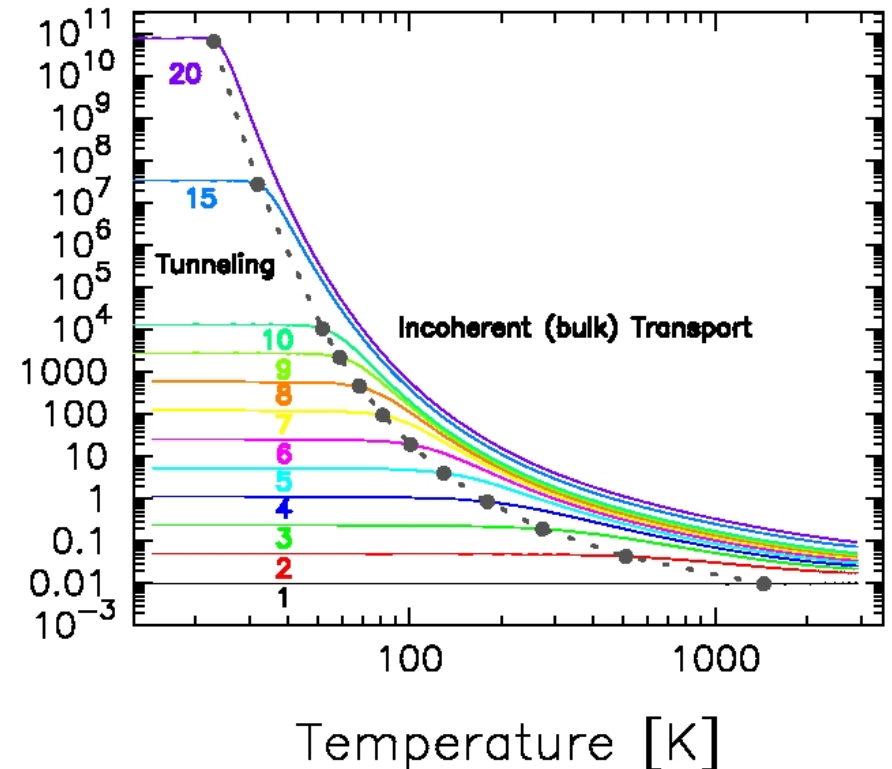
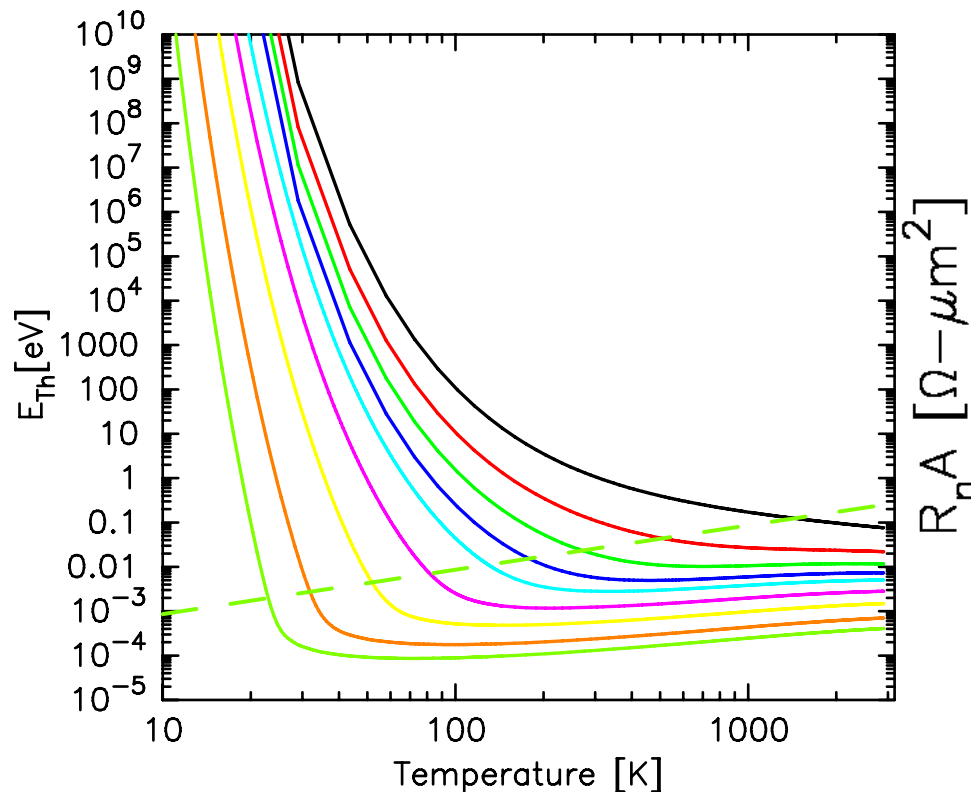
- Generalizing the above relation to an insulator by

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

gives the general form for the Thouless energy.

Temperature dependence

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!

Vertex corrections ...

Irreducible Charge Vertex

- In the Falicov-Kimball model, the irreducible charge vertex for dynamical charge responses is known

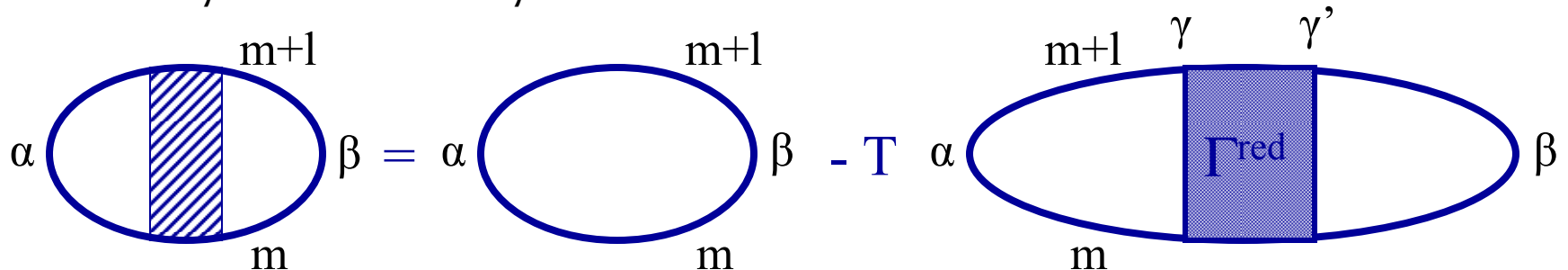
$$\Gamma_{\alpha}^{irred}(i\omega_m, i\omega_n; i\nu_l) = \delta_{mn} \frac{1}{T} \frac{\Sigma_{\alpha}(i\omega_m) - \Sigma_{\alpha}(i\omega_{m+l})}{G_{\alpha}(i\omega_m) - G_{\alpha}(i\omega_{m+l})}$$

- Hence one can solve for the isothermal conductivity matrix directly by solving the relevant Bethe-Salpeter-like equations.

Schematic picture for the polarization

- The polarization response can be represented pictorially in terms of the reducible charge vertex matrix.

$$\Gamma_{\gamma\gamma'}^{red} = \Gamma_{\gamma}^{irred} \left[1 + \sum_{k^{\parallel}} G_{\alpha\beta}(k^{\parallel}, i\omega_m) G_{\beta\alpha}(k^{\parallel}, i\omega_{m+l}) \right. \\ \left. \times \frac{\Sigma_{\beta}(i\omega_m) - \Sigma_{\beta}(i\omega_{m+l})}{G_{\beta}(i\omega_m) - G_{\beta}(i\omega_{m+l})} \right]^{-1}_{\gamma\gamma'}$$

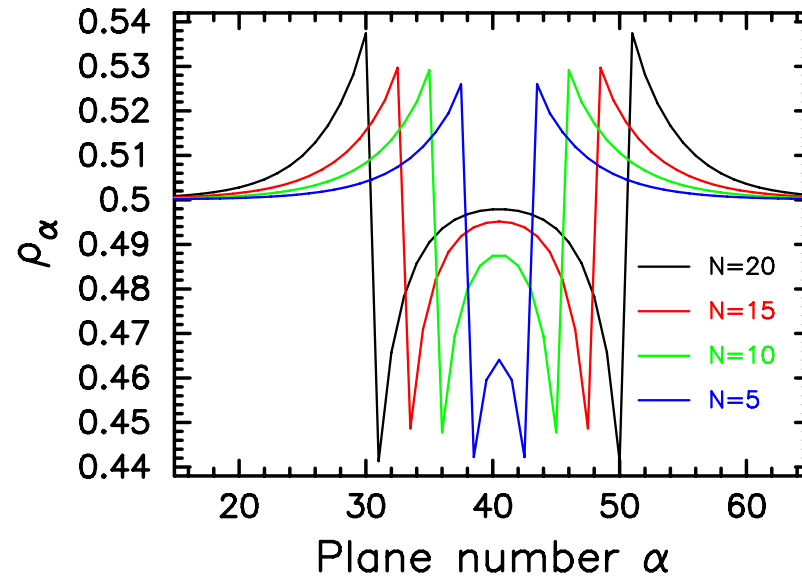
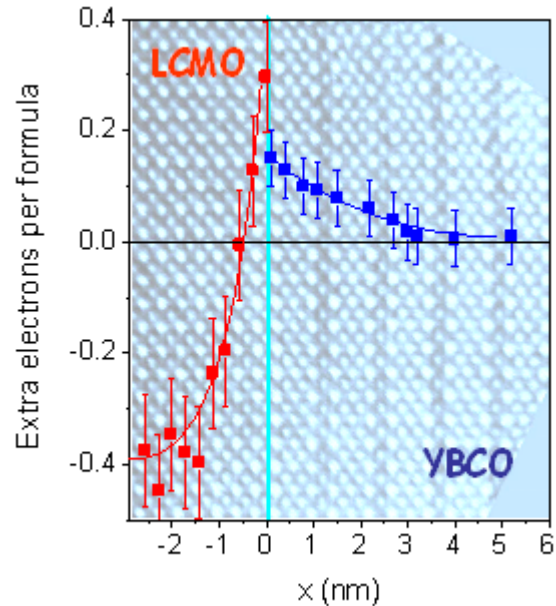


Finishing the calculation

- One needs to analytically continue the result from the imaginary to the real-frequency axis. This is straightforward to do if we assume the matrix in the reducible charge vertex does not introduce any poles in the UHP.
- Of course the matrix needs to be truncated to a finite size, but we include all correlated planes and the region around the interface to pick up the most important terms.
- We expect to have actual results from these calculations soon...

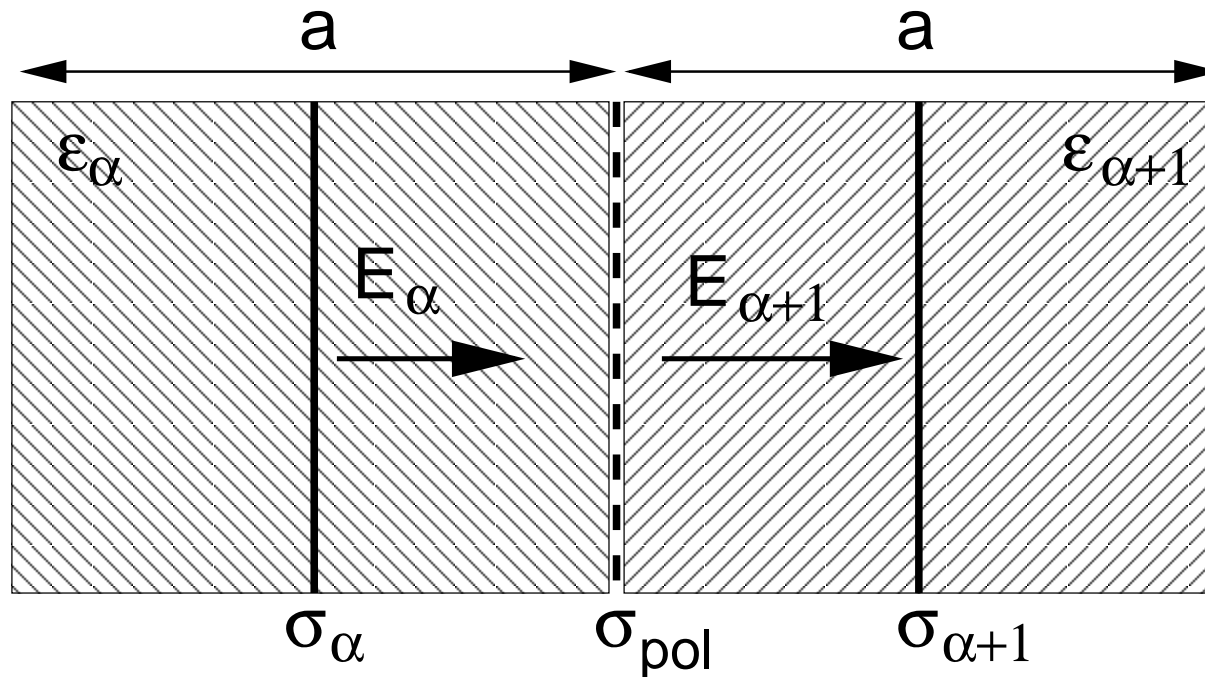
Particle-hole asymmetry is
necessary for thermoelectric
devices ...

Electronic charge reconstruction



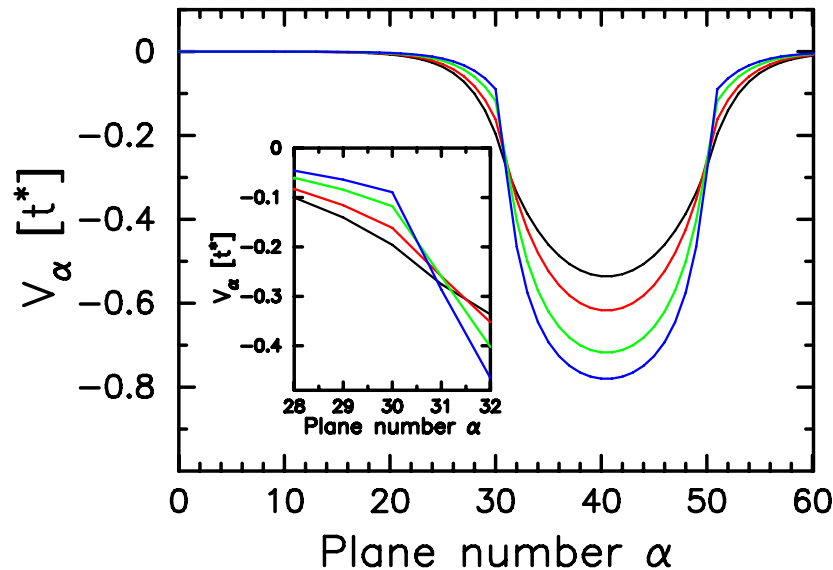
Using a scanning transmission electron microscope with electron energy-loss spectroscopy, one can directly measure the electronic charge at each plane of a strongly correlated multilayered nanostructure. Left are experimental results by Varela et al. on YBCO/LCMO heterostructures, right is a simple theory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

Theoretical treatment

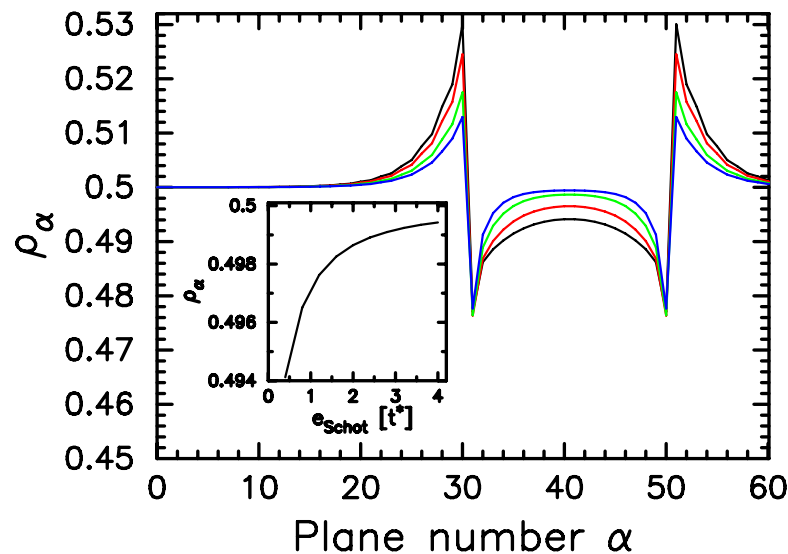


We employ a semiclassical treatment to handle the electronic charge reconstruction. We allow charge to be rearranged on different planes, as determined by the electrochemical potential at a given plane site, and then determine the classical Coulomb potential from planes of net charge, with dielectric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

Coulomb potential

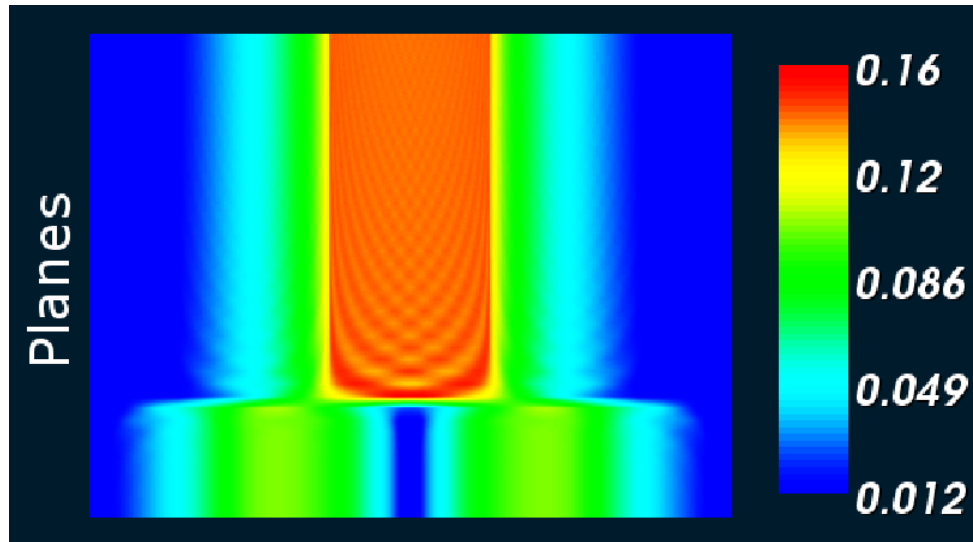


The Coulomb potential develops a kink at locations where the dielectric constant changes (i.e. at the interfaces), and it goes to zero far from the interface due to overall conservation of charge.

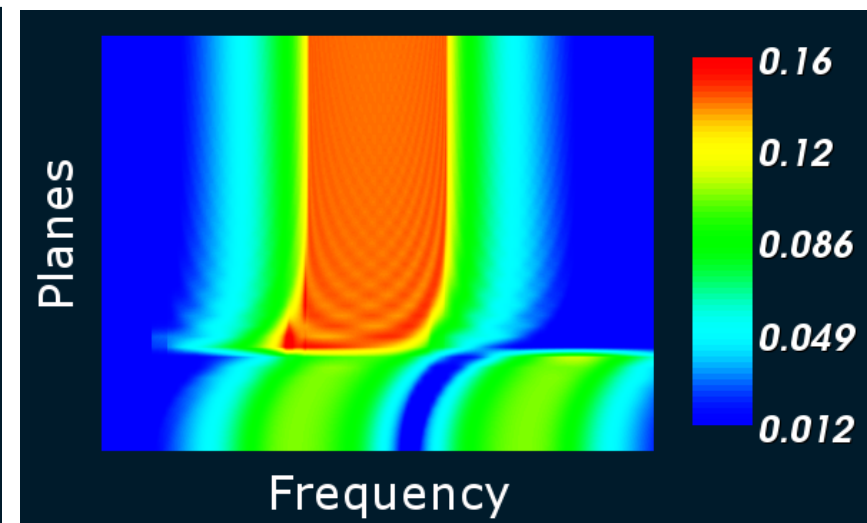
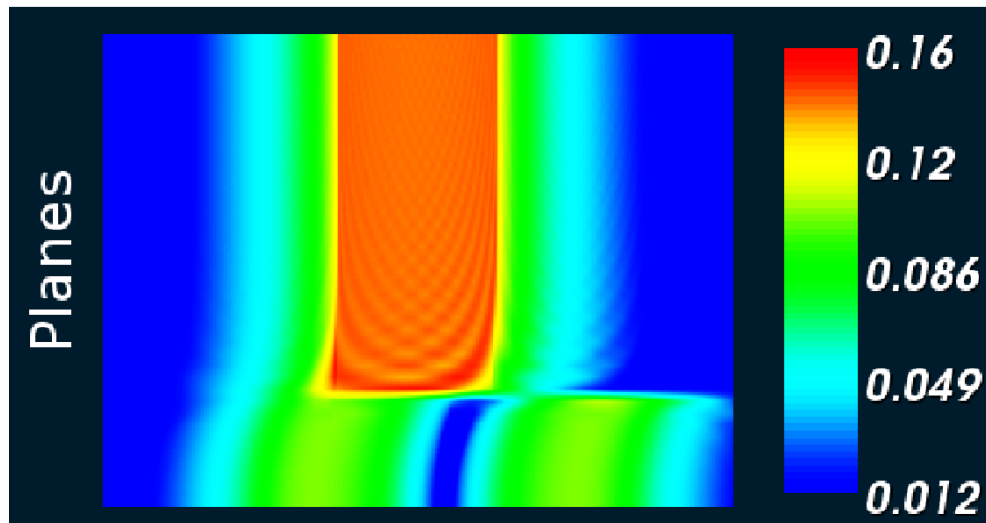


As the screening length decreases, the total charge that is rearranged gets smaller for a fixed chemical potential mismatch of the bulk materials.

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:

$$j_{\alpha} = e^2 \sum_{\beta} L^{11}_{\alpha\beta} E_{\beta} - e \sum_{\beta} L^{12}_{\alpha\beta} (T_{\beta+1} - T_{\beta-1}) / 2a$$

$$j_{Q\alpha} = \sum_{\beta} L^{21}_{\alpha\beta} E_{\beta} - \sum_{\beta} L^{22}_{\alpha\beta} (T_{\beta+1} - T_{\beta-1}) / 2a$$

Where the L matrices are found from the **Jonson-Mahan theorem** (current and heat-current correlation functions in real space) which holds even with vertex corrections.

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 $E_{\alpha} = \sum_{\beta\gamma} (L^{11})^{-1}_{\alpha\beta} L^{12}_{\beta\gamma} (T_{\gamma+1} - T_{\gamma-1}) / 2a$

The temperature gradients become

$$\sum_{\beta} M^{-1}_{\alpha\beta} j_Q = -(T_{\alpha+1} - T_{\alpha-1}) / 2a; \quad M = -L^{21} (L^{11})^{-1} L^{12} + L^{22}$$

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

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

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!**

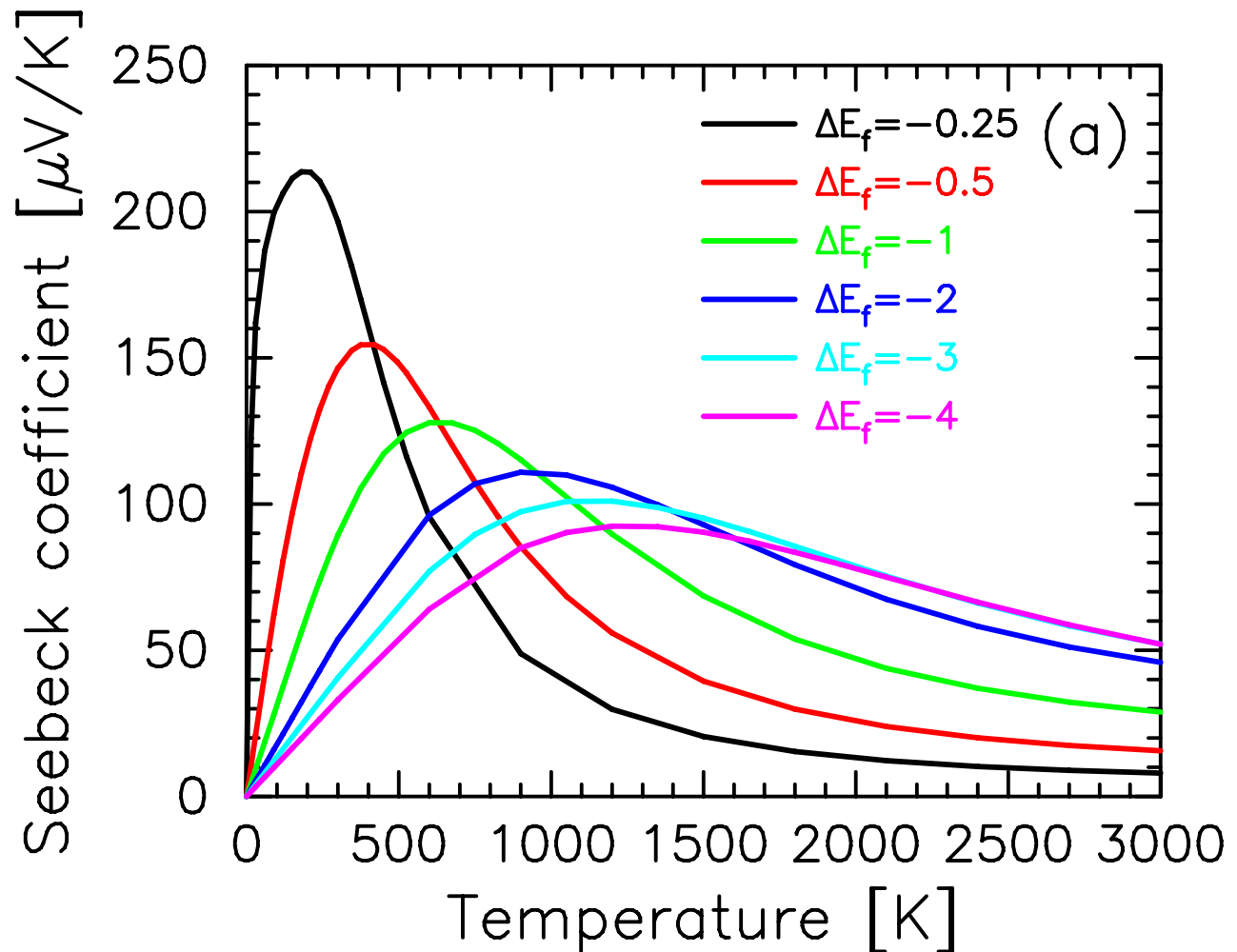
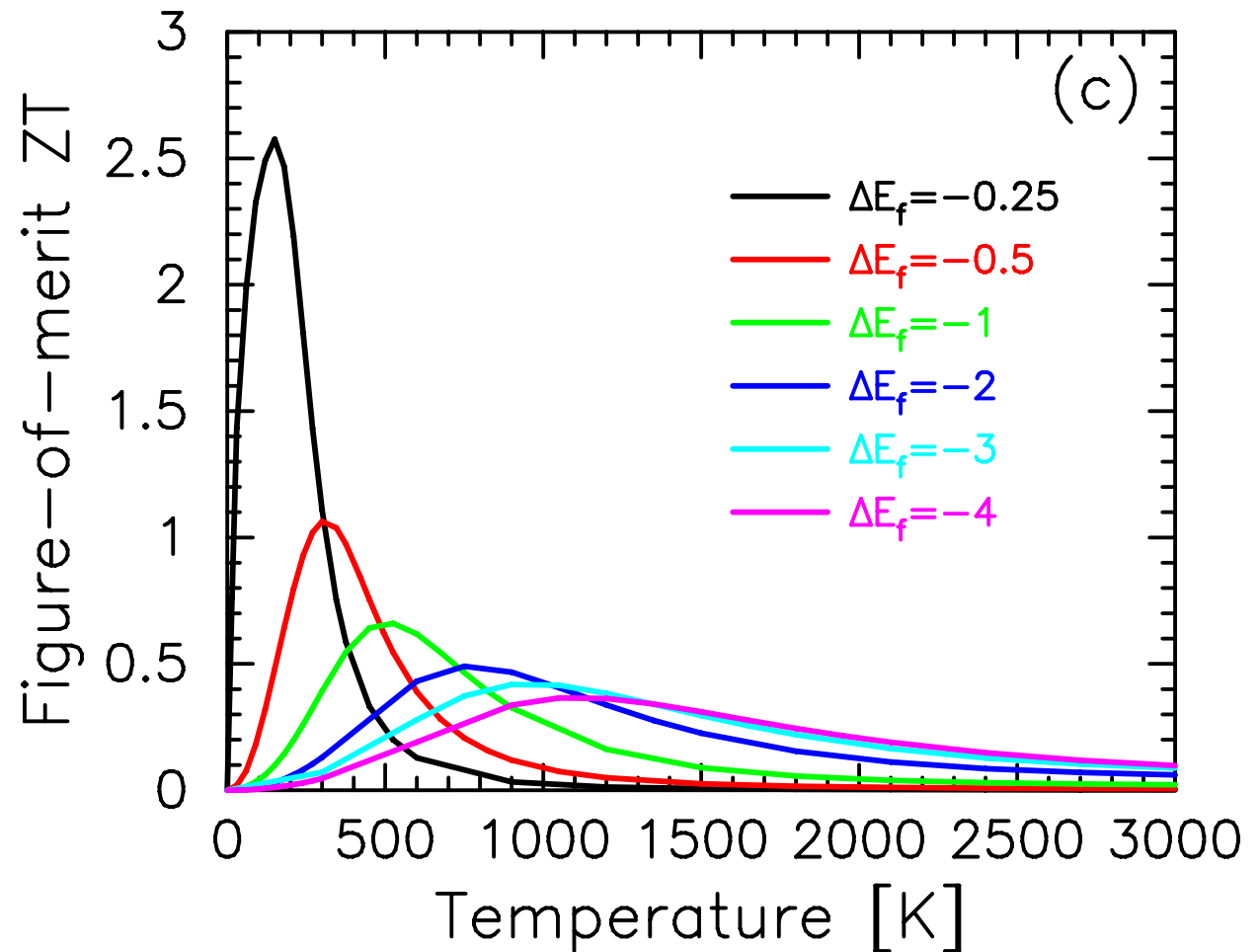


Figure of merit

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.



Creation of a low temperature conducting channel with a Mott insulator described by the Hubbard model

Fermi-liquid behavior

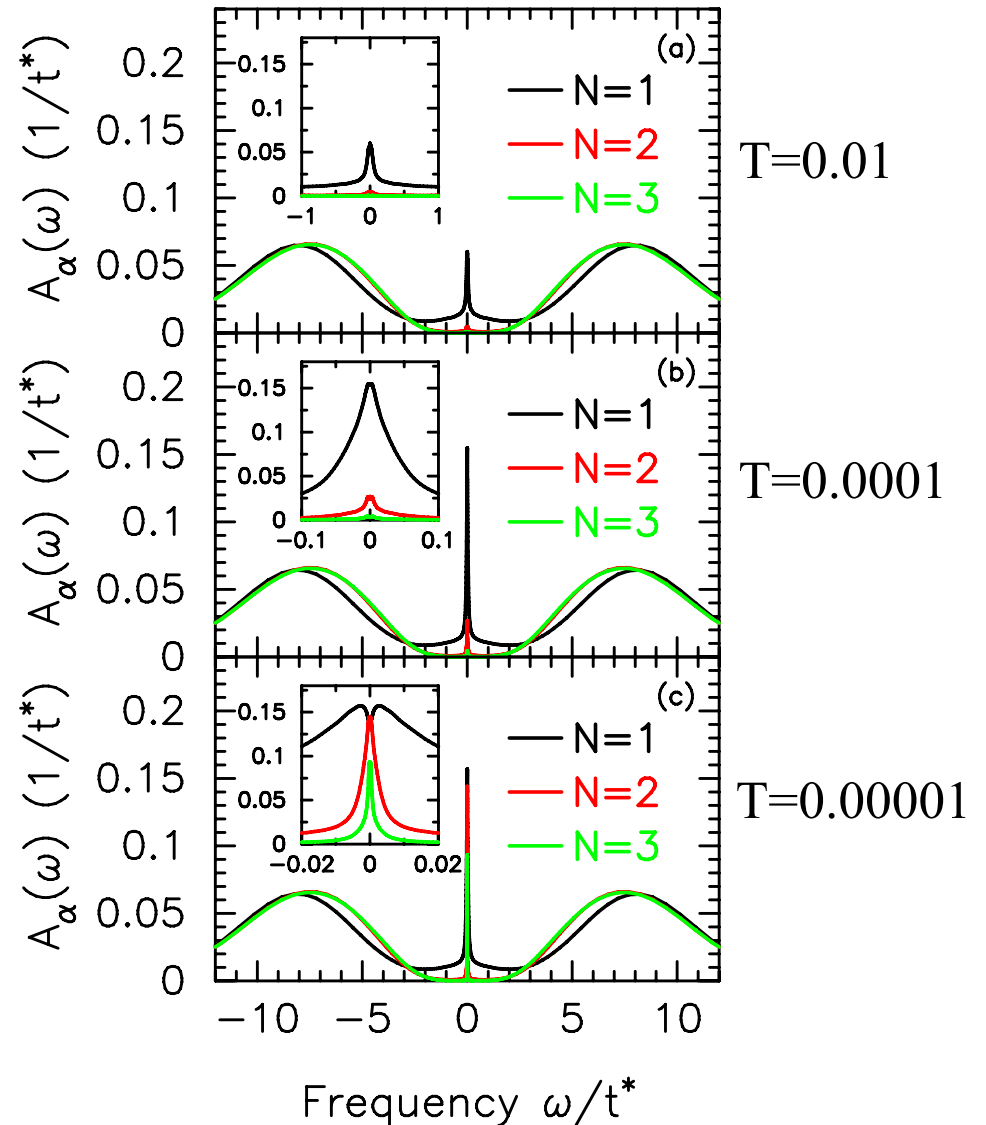
- The Hubbard model shows FL behavior in the metallic phase. When we examine a finite sandwich of a Mott insulator between two semi-infinite metallic leads, the normal state proximity effect creates a tunneling link between the metals, which implies that the renormalized hybridization for the SIAM on each plane never goes to zero.
- *Hence, at $T=0$, a FL forms throughout the device, and it becomes perfectly conducting!*

Quantum emergent phenomena at $T=0$

- Even though the system is inhomogeneous, at $T=0$, **because the self-energy vanishes on all of the Hubbard planes**, the system restores periodicity, has a well-defined Fermi surface, and has the noninteracting DOS at zero frequency.
- *This state will be very fragile to finite T , frequency, voltage, or disorder, but for thin barriers close to the metal-insulator transition it should be possible to observe it.*

Example: $N=5$ barrier with $U=16$

- The T_F for each plane gets smaller as we move deeper into the barrier, so the coherence peak develops at different temperatures, which should give rise to highly nonlinear behavior in the current-voltage characteristics.
- *For more details, please see Hand Zenia's talk!*



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; (iv) thermal transport, the Seebeck effect, and the figure-of-merit; and (v) the opening of a perfect conducting channel in a metal-Mott-insulator-metal device at $T=0$.

The interplay between inhomogeneity and electron-electron correlations leads to interesting new phenomena, that could have wide-ranging applications. We have only begun to see where this work will ultimately lead.