Many body physics issues with the core-hole propagator and resonant inelastic X-ray scattering

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Core-hole creation

- In second-order X-ray scattering processes, a high-energy photon is absorbed by an ion which excites a core electron to the vicinity of the Fermi level.
- A core hole is left behind, which has a locally modified Coulomb interaction with the remaining electrons, causing them to relax.
- When an excited electron drops down to fill the core-hole state, light with changed momentum and energy emerges, and is measured.

Equilibrium or nonequilibrium?

The creation of the core-hole is an inherently local and nonequilibrium process, since the core hole is created at a specific time and place.

But, if we assume the conduction electrons provide an adiabatic bath for the core-hole, or if we assume the conduction electrons instantaneously change to a new inhomogeneous steady state, then an equilibrium formulation can be used. Otherwise, a nonequilibrium approach is needed to self-consistently determine how the system switches to the inhomogeneous steady state after the core-hole is created.

Core-hole propagator

The core-hole propagator is a complicated object. It is associated with the so-called orthogonality catastrophe in near-edge X-ray absorption studies, where the propagator diverges with a power law related to the Coulomb interaction between the core hole and the conduction electrons in metals. In RIXS studies, one often assumes it has a constant lifetime determined by Auger and other nonradiative decay processes.

Approximate treatments

Delta function in time (instantaneous)

Delta function in frequency (static)

Noninteracting form with a constant relaxation time entered by hand

Local approximation for the impurity problem (without self-consistency with the lattice), but with local dynamics treated exactly, ignoring all nonradiative decay channels; also one can solve the inhomogeneous impurity problem for the case of fast conduction electron relaxation.

Falicov-Kimball-like interaction

The core hole is completely localized, so it has no hybridization with neighboring lattice sites. It interacts with other electrons entirely via its screened Coulomb interaction Q.

$$H_{core-hole} = (E_c + Q n_{conduction}) n_{core-hole}$$

Different Q values can be associated with different electrons near the chemical potential.

Quantum-mechanical formalism

The core-hole number operator is a conserved quantity of the Hamiltonian, so it can be replaced by a classical value corresponding to its value in each term of a trace over quantum-mechanical states. This makes the core-hole interaction with conduction electrons into a “quasi-quadratic” interaction, which can be solved immediately by the Feynman fermionic determinant.

Dynamical mean-field theory

The DMFT approach maps the lattice problem onto an equivalent impurity problem in time-dependent fields, which mimic the hopping of electrons.

The core-hole electron propagates in this effective DMF which involves a new impurity problem that must be solved.

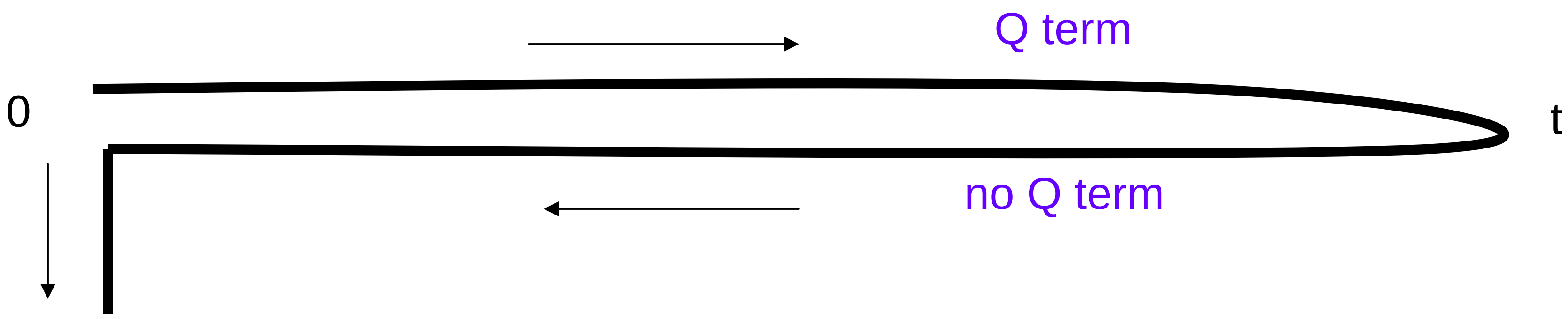
We must use a modified impurity solver for the core-hole propagator from that used for the correlated system (NRG or Kadanoff-Baym-Keldysh approach).

NRG approach

In the NRG approach, we map the time-dependent field onto a series of discrete delta functions, and then create a one-dimensional auxiliary problem that mimics the coupling to the time-dependent fields by couplings in space. This algorithm can be straightforwardly modified to handle the calculation of the core-hole propagator, but is has numerical issues at large frequencies.

Keldysh-Kadanoff-Baym approach

By using an equation of motion, the core-hole Green's function can be expressed as the average of a complicated time-ordered-product, with an additional factor due to the Q interaction that is not time-translation invariant on the contour.

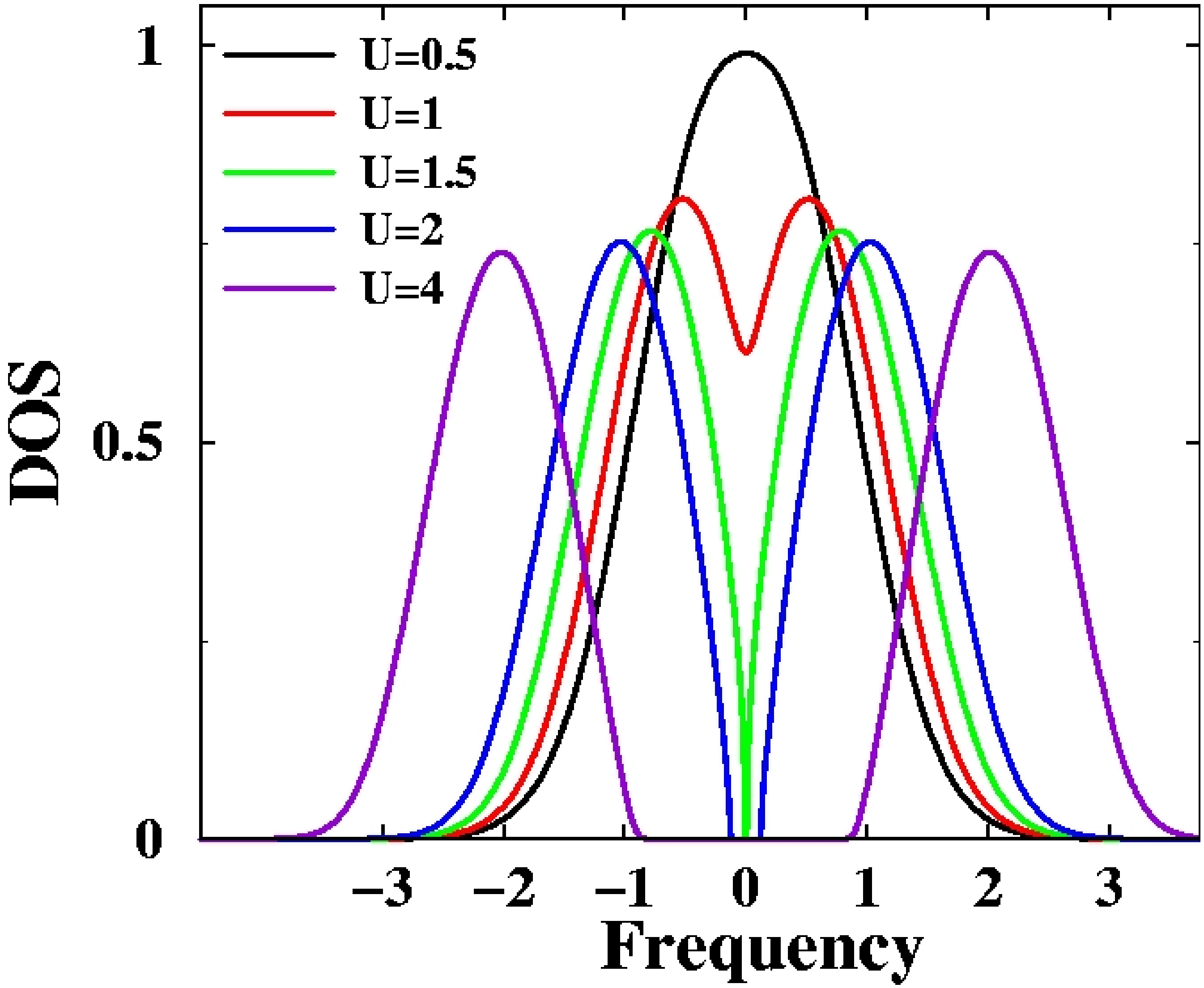


Falicov-Kimball model

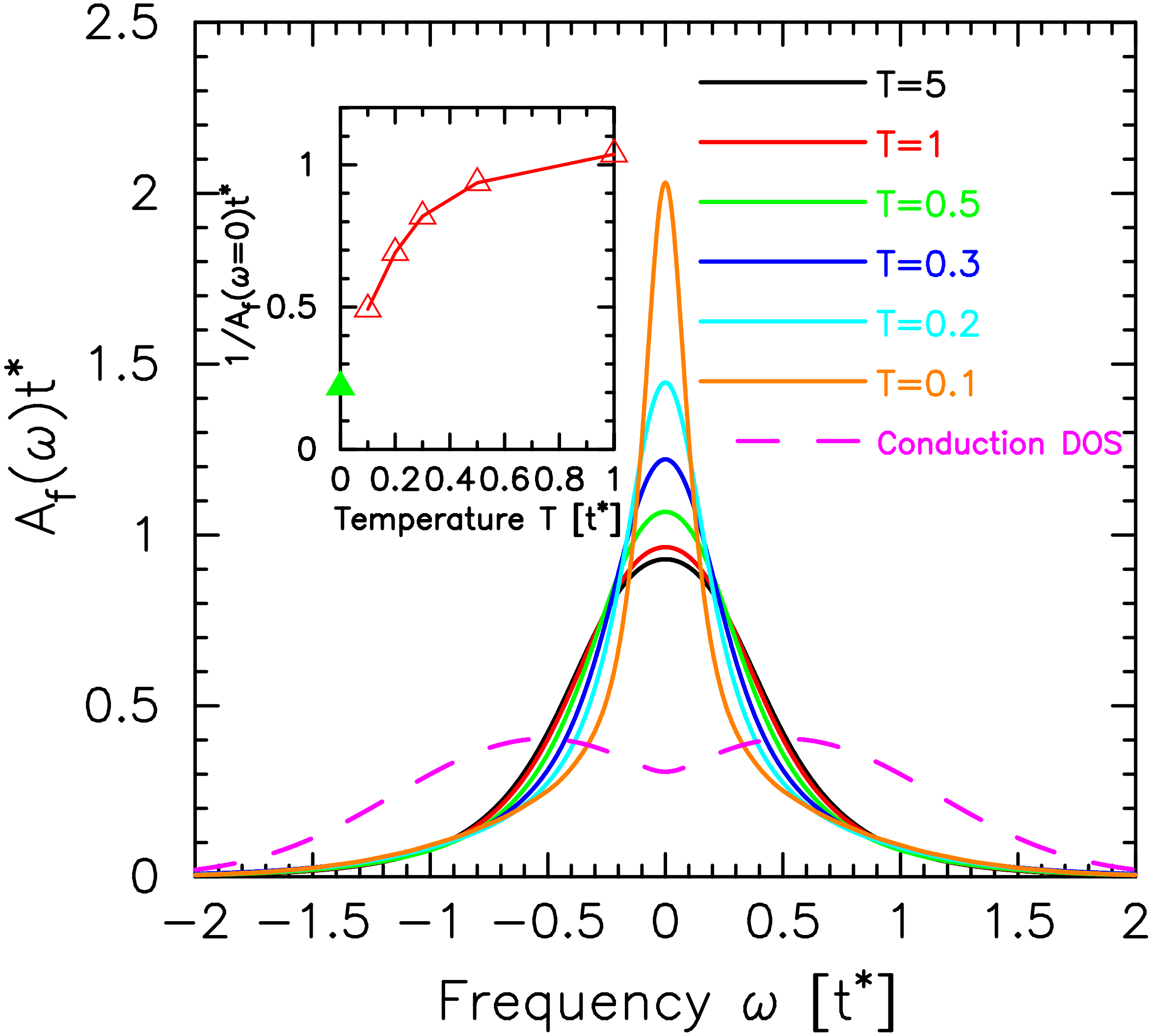
In this model, we have mobile d electrons and static f-electrons at the fermi energy. They interact with each other with an energy U, which can drive the system into an insulating state for $U>1.414$.

We examine the local core-hole propagator when $U=1$ (metal) and $Q_d=1$, $Q_f=0$

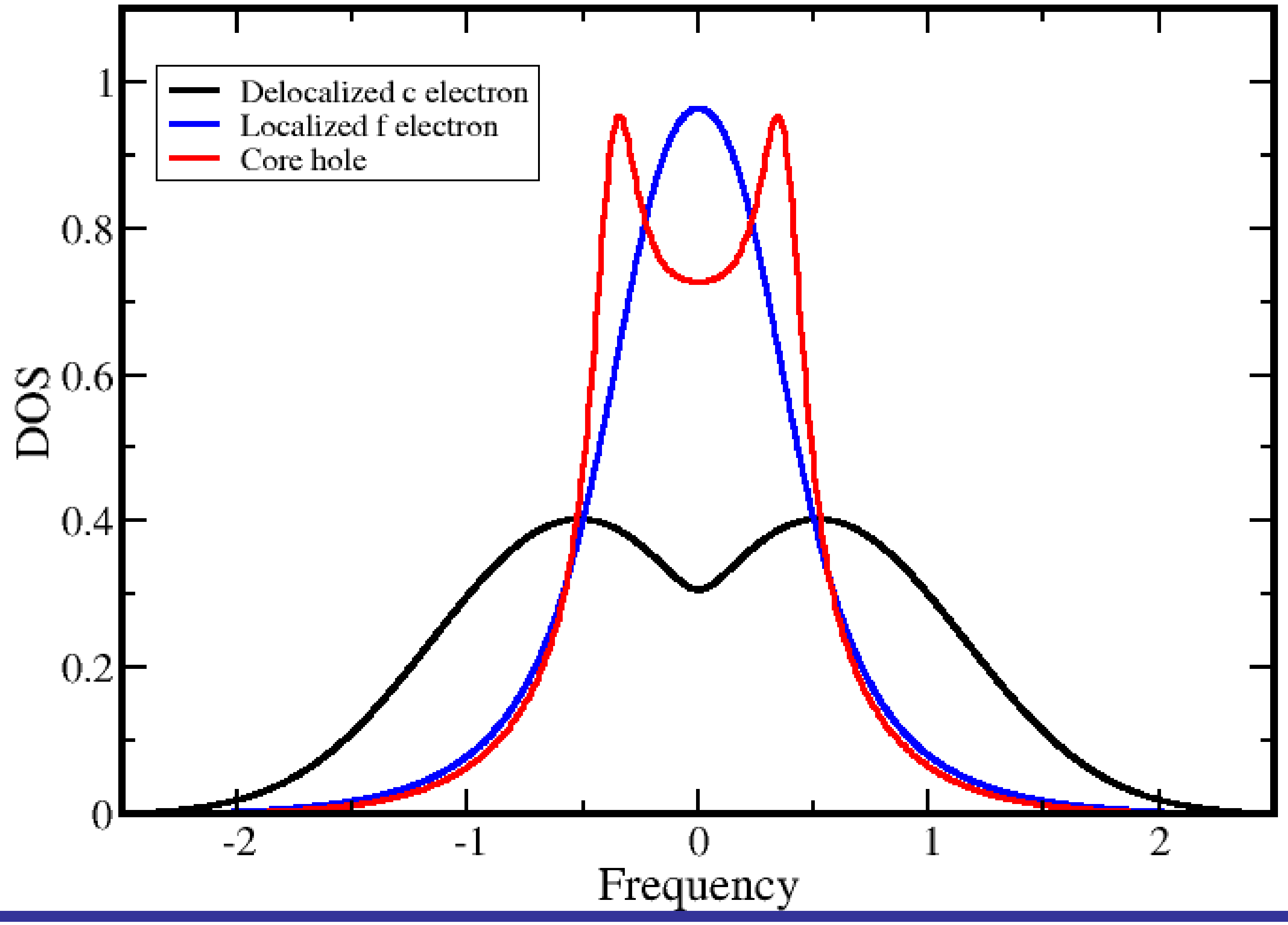
MIT in Falicov-Kimball model



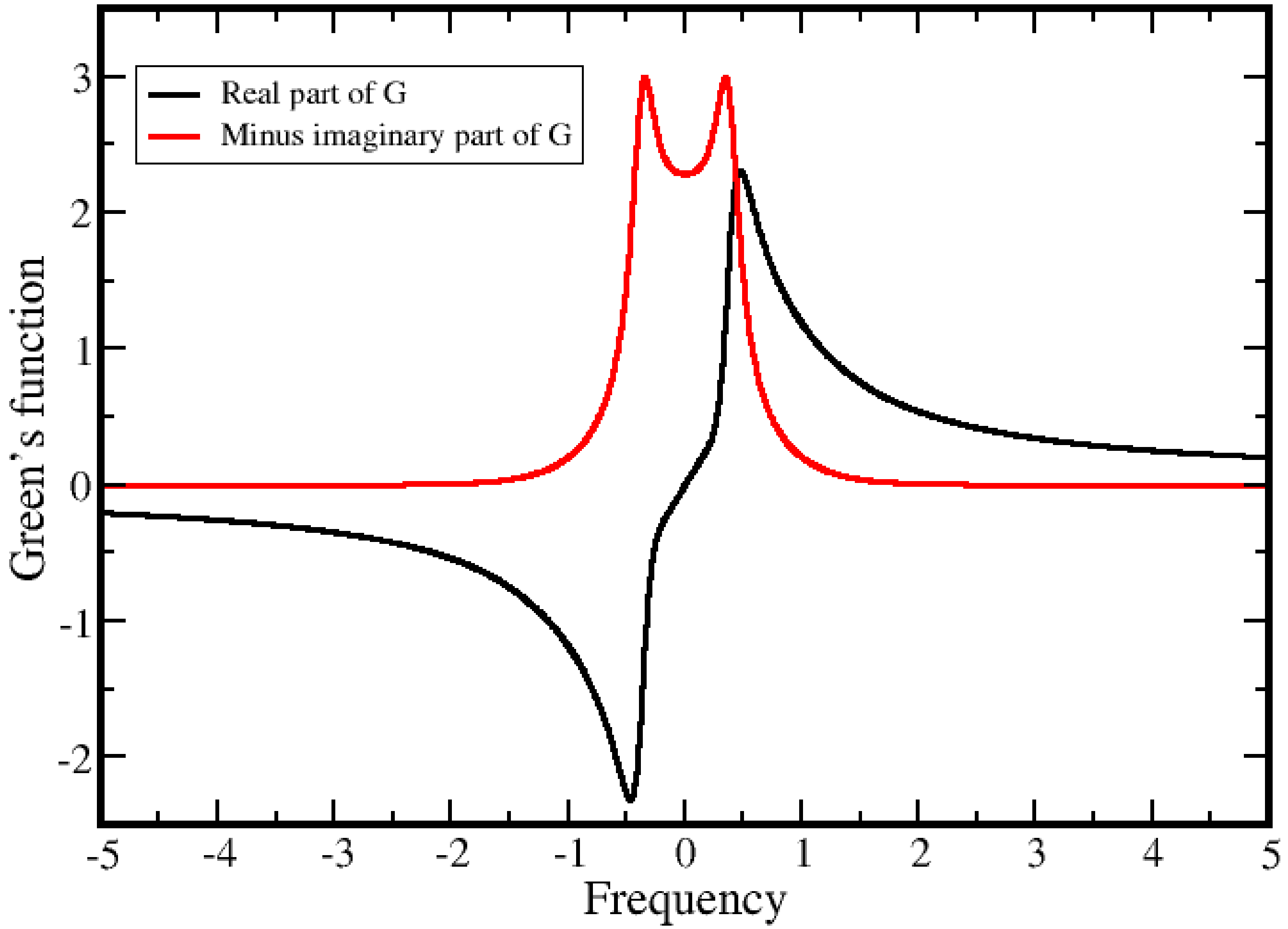
F-electron DOS



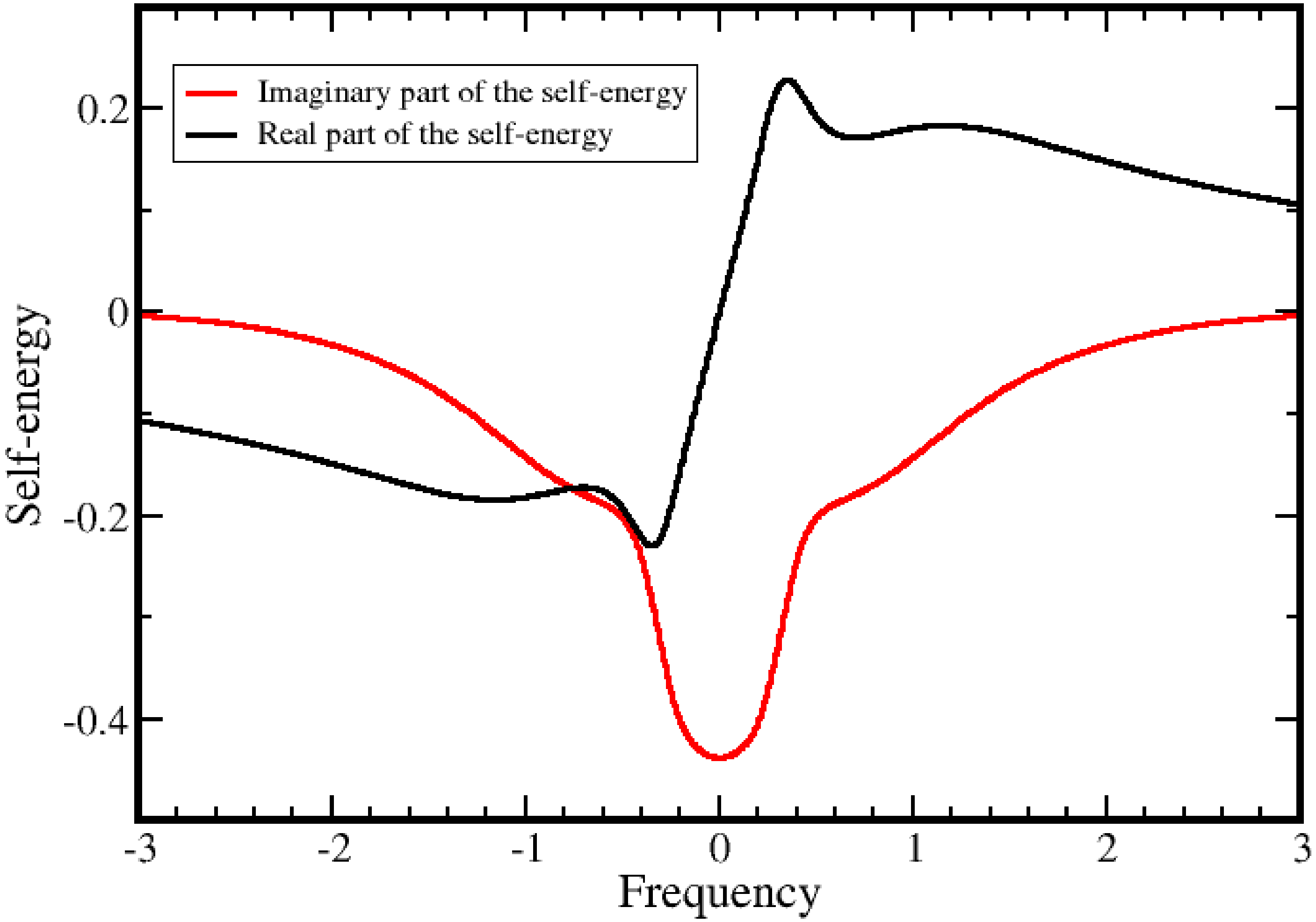
DOS for U=1, Q_d=1, Q_f=0



Core-hole propagator



Core-hole self-energy



Implications for approximations

Whenever the imaginary part of the self-energy is larger than the inverse of the Auger lifetime, the many-body correlations determine the shape of the propagator, not the Auger lifetime. In this case, the self-energy dominates for lifetimes longer than about 1-10 fs. The many-body correlations become more important as the interactions increase.

RIXS

The RIXS cross section involves the core-hole propagator, the propagator of electrons near the fermi energy, and two-particle charge-screening processes.

Inevitably the screening process will need to be handled in an approximate way, but for some models one can evaluate many of the important diagrams exactly (within DMFT)

Future plans

Complete the exact analysis of core-hole propagator for the Falicov-Kimball model and apply it to solving for different RIXS scattering processes

Develop NRG approaches for more complicated many-body problems like the Hubbard model

Introduce more materials specificity to the approach

Expertise

We have expertise in DMFT techniques for determining single-particle properties such as the propagators of all electrons in the system and in two-particle properties related to screening effects.

Our expertise spans NRG techniques, Keldysh-Kadanoff-Baym approaches, and Bethe-Salpeter and more complicated screening processes.

Collaborations

Need expertise in atomic physics for examining multiplet structures/effects and polarization effects

Need DFT expertise to determine relevant band-structures and mappings to effective tight-binding models.

Need experimental input to determine what the most important problems/questions are to examine.