ELECTRONIC CORRELATIONS AND TRANSPORT PROPERTIES OF MOLECULAR METALS: A DYNAMICAL MEAN-FIELD TREATMENT

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MAIN POINTS

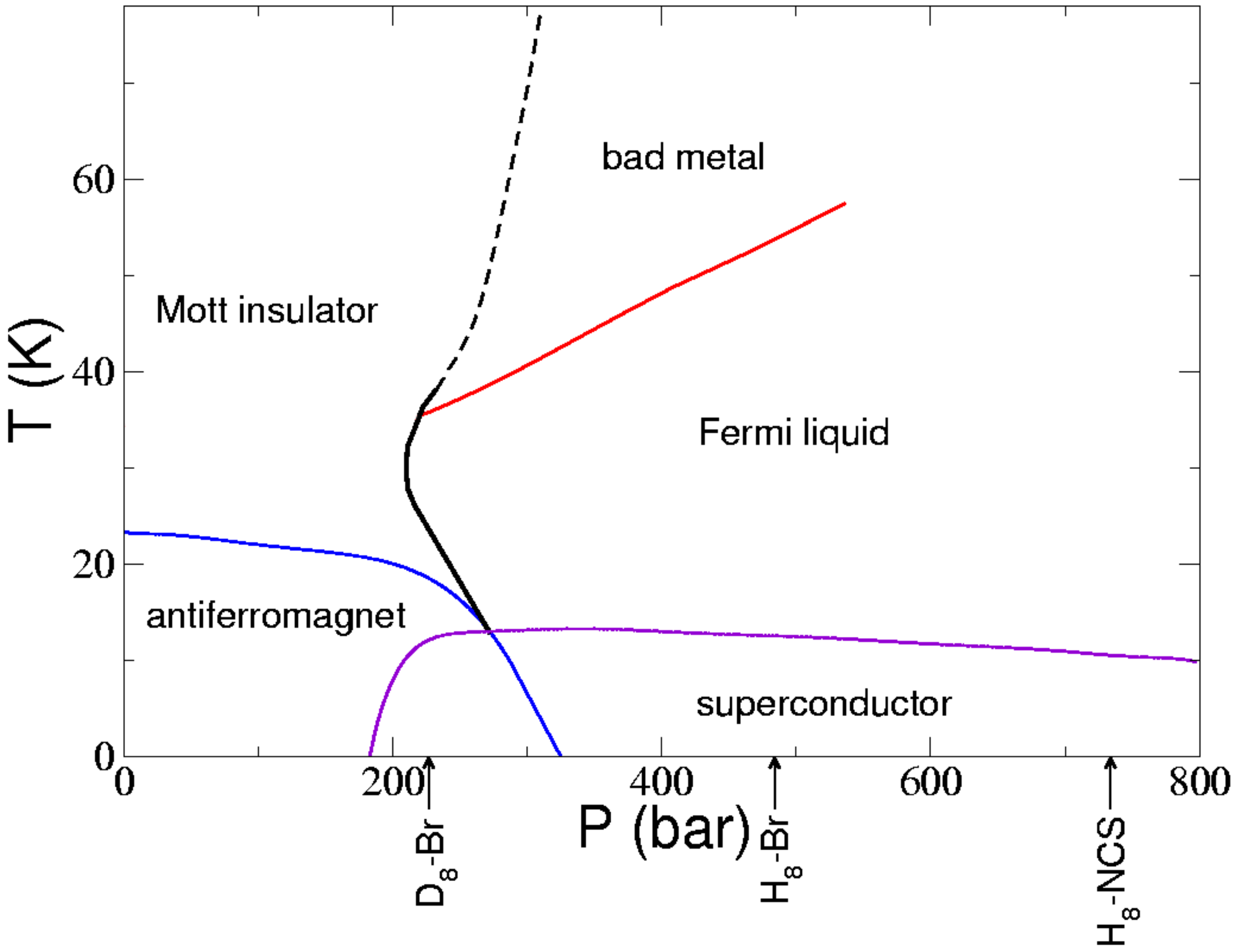
(1) The metallic phase of κ-(BEDT-TTF)$_2$X is a strongly correlated metal close to a Mott insulating phase. It exhibits a crossover from a Fermi liquid to a bad metal at $T_0 \sim 30$ K.

(2) The relevant theoretical model is a Hubbard model on an anisotropic triangular lattice at half-filling.

(3) Transport properties, including the coherence temperature T_0 is described by dynamical mean-field theory.

κ-(BEDT-TTF)$_2$X PHASE DIAGRAM

Temperature vs. pressure



- metal-insulator transition is first order
- bad metal near metal-insulator transition
- competition between superconductivity and antiferromagnetism
- similar to cuprates except pressure plays the role of doping

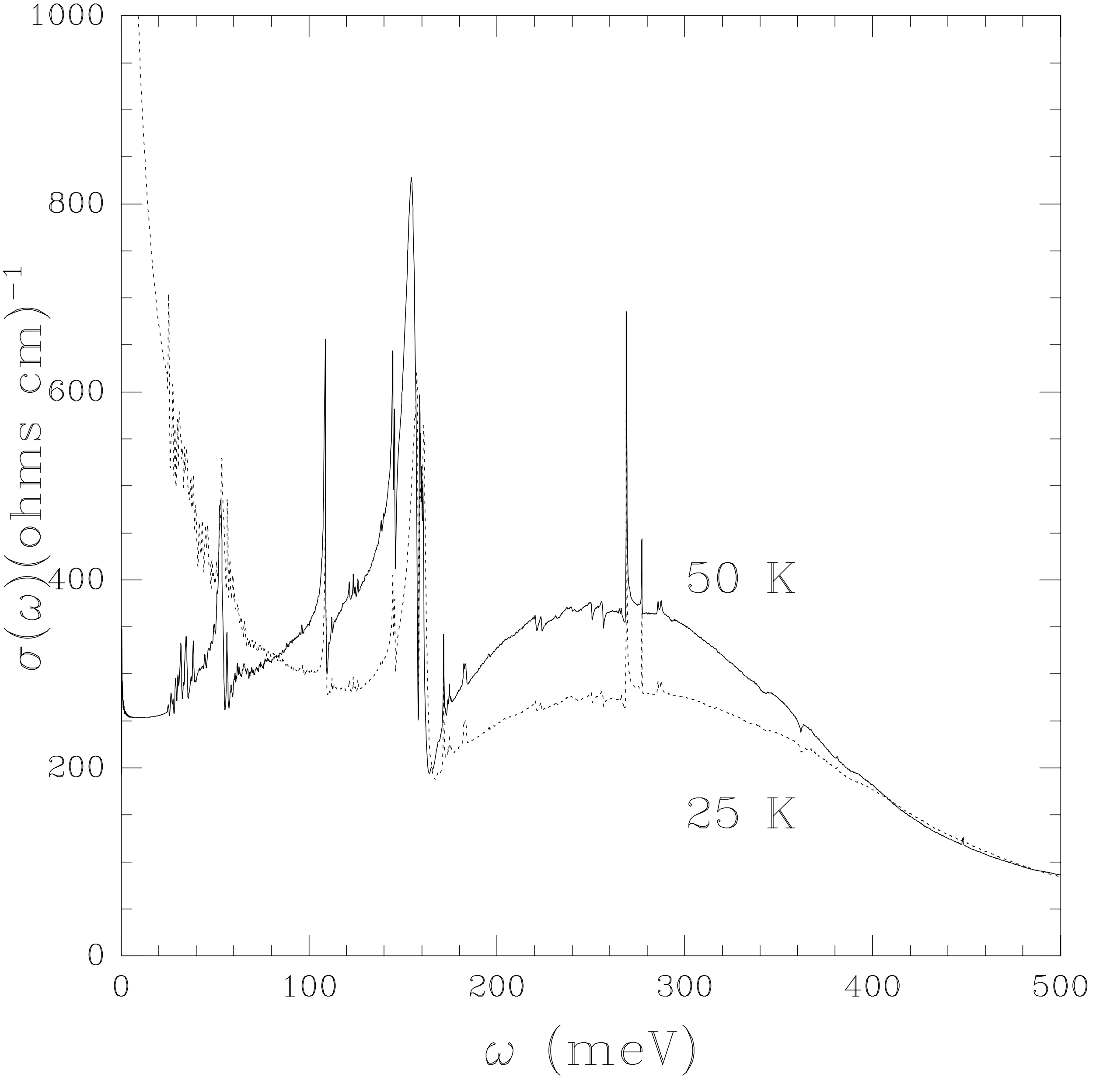
UNCONVENTIONAL METALLIC PROPERTIES

Temperature dependence of resistivity

- $\rho(T)$ does not monotonically increase with temperature
- $\rho \sim T^2$ at low temperatures as for a Fermi liquid
- above 30 K the “mean-free path” is much less than a lattice constant (i.e., a bad metal).

UNCONVENTIONAL METALLIC PROPERTIES

Temperature dependence of optical conductivity



- No Drude ($\omega = 0$) peak above $T_0 \sim 50$ K
- Broad peak around 300 meV.

UNCONVENTIONAL METALLIC PROPERTIES

Temperature dependence of thermopower

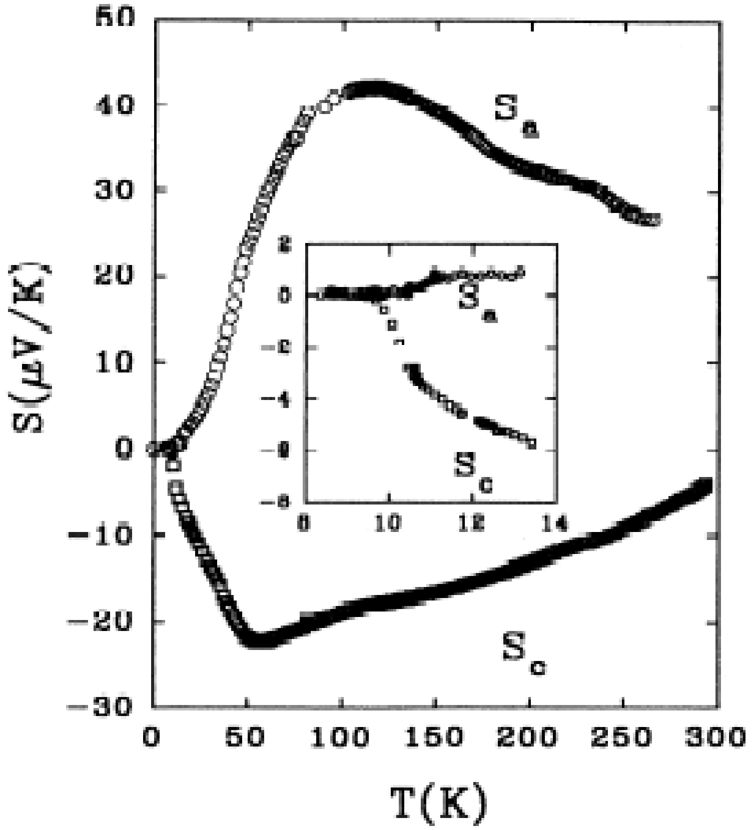


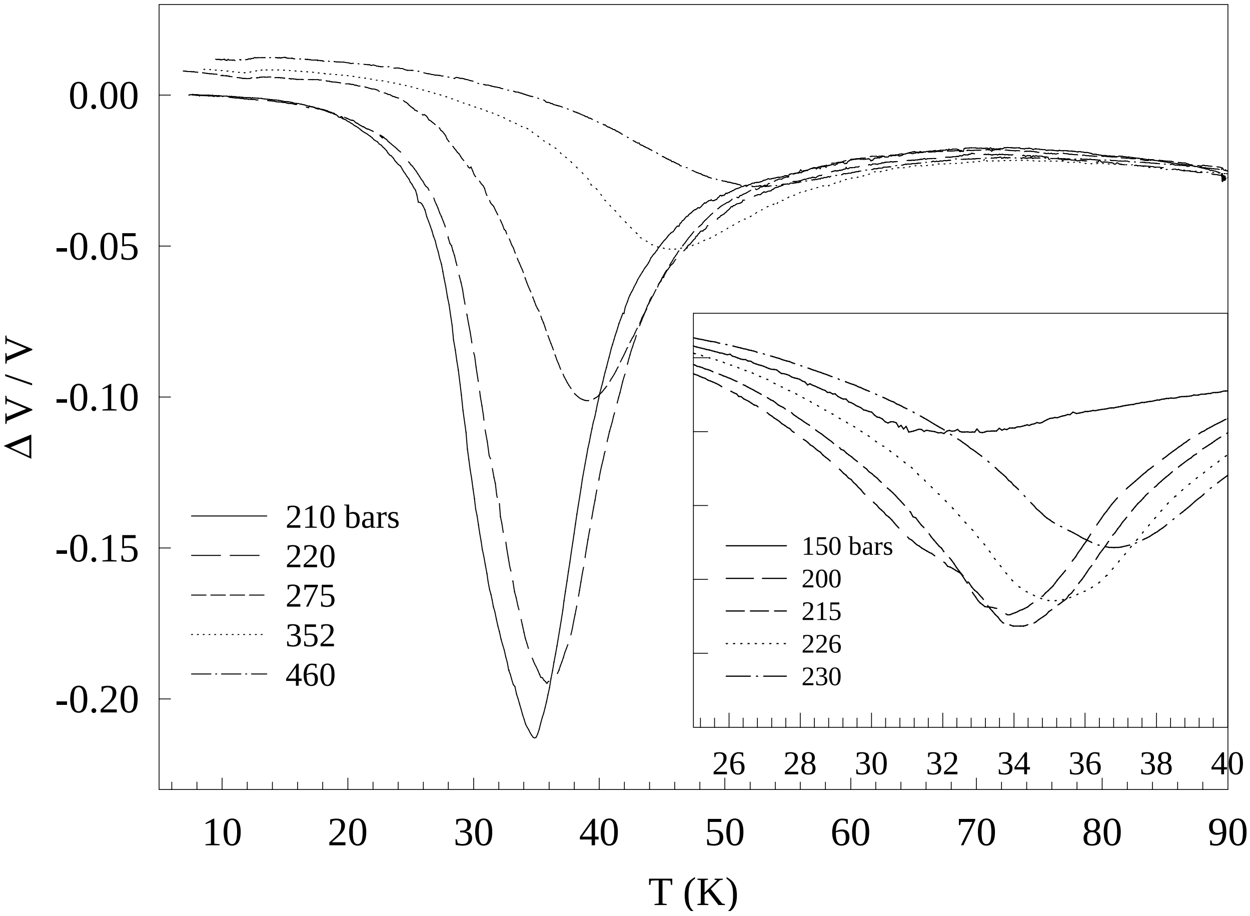
FIG. 2. Observed temperature dependence of the thermopower of κ-(BEDT-TFF)$_2$Cu[N(CN)$_2$]Br in the a and c directions from 300 to 4 K. In the inset we show the data near $T_c \sim 10.5$ K.

- Values of order of $k_B/e \sim 76$ μV/K
- Non-monotonic temperature dependence

UNCONVENTIONAL METALLIC PROPERTIES

Temperature dependence of speed of sound

Fournier *et al.*, PRL **90**, 127002 (2003).



- Anomaly at T_0 is orders of magnitude larger than at superconducting transition.
- Anomaly increases closer to the Mott insulator.

CONVENTIONAL METALLIC PROPERTIES

Quantum magnetic oscillations

The temperature and magnetic field dependence of SdH and dHvA oscillations is consistent with a 2d or quasi-2d Fermi liquid at low temperatures ($T < 3$ K).

$$\frac{m^*}{m_e} \sim 1 - 7 \qquad\qquad \frac{\hbar}{\tau\ k_B} \sim 0.1 - 2 \text{ K}$$

Merino and RHM, *PRB* **62**, 2416 (2000).

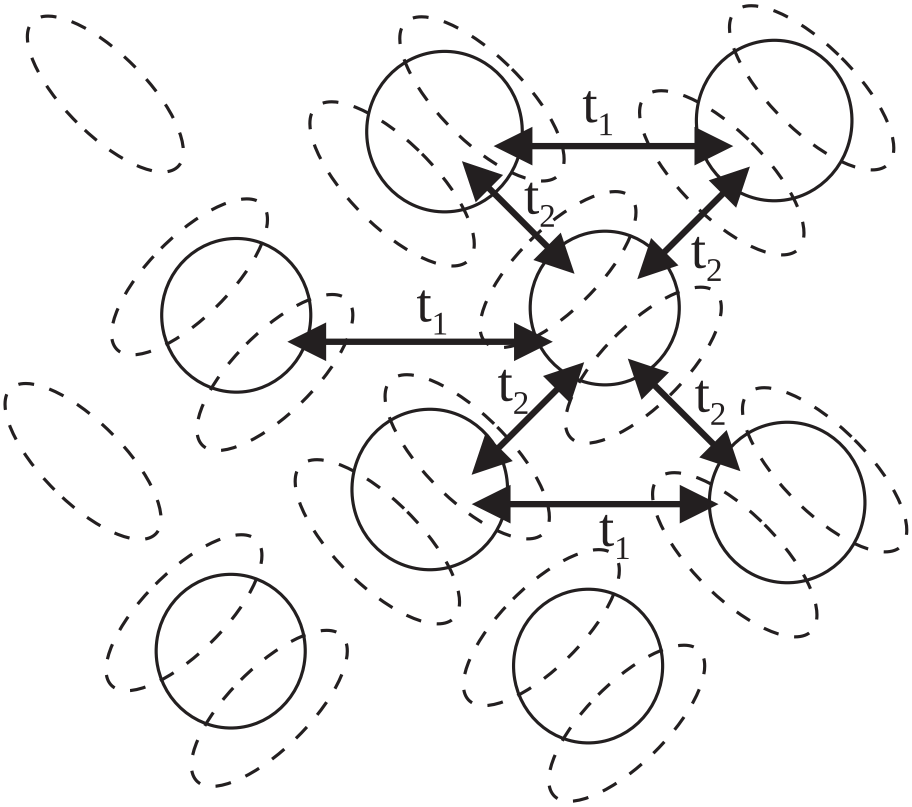
CONVENTIONAL METALLIC PROPERTIES

Angle-dependent magnetoresistance oscillations

The angle dependence is consistent with a 2d or quasi-2d Fermi liquid with semi-classical quasi-particle dynamics and can be used to map out the Fermi surface.

THE RELEVANT HUBBARD MODEL FOR κ-(BEDT-TTF)$_2$X

RHM, *Comments CMP* **18**, 309 (1998).



There is one hole per dimer.

The dimers form an *anisotropic triangular lattice* with different hopping in the horizontal (t_1) and slanted directions (t_2).

U is the Coulomb repulsion between two holes on a dimer. From quantum chemistry U is larger than the band width.

This is a strongly correlated system.

Magnetic frustration results from competition between t_1 and t_2.

THE RELEVANT HUBBARD MODEL

$$\begin{aligned} H = & - t_1 \sum_{<\mathbf{ij}>,\sigma} (c^\dagger_{\mathbf{i},\sigma} c_{\mathbf{j},\sigma} + h.c.) \\ & -t_2 \sum_{<\mathbf{in}>,\sigma} (c^\dagger_{\mathbf{i},\sigma} c_{\mathbf{n},\sigma} + h.c.) \\ & +U \sum_{\mathbf{i}} (n_{\mathbf{i}\uparrow} - \frac{1}{2})(n_{\mathbf{i}\downarrow} - \frac{1}{2}) + \mu \sum_{\mathbf{i},\sigma} n_{\mathbf{i}\sigma} \end{aligned}$$

At half filling (one electron per site) ground state is an *insulator* for $U \gg t_1, t_2$.

If t_1 and t_2 are both non-zero then due to imperfect nesting of the Fermi surface there is a Mott-Hubbard *metal-insulator transition* at a finite value of U.

DYNAMICAL MEAN-FIELD THEORY OF THE MOTT-HUBBARD TRANSITION

A. Georges *et al.*, RMP **68**, 13 (1996).

- DMFT treats temporal fluctuations on a single lattice site exactly but neglects spatial correlations.
- It becomes exact in the limit of large lattice connectivity or spatial dimensionality.
- It maps the Hubbard model onto a single impurity problem (Anderson model) that is solved self-consistently.

- Leads to a new low energy scale $T_0 \ll U$ and D (the band width).
  T_0 corresponds to the Kondo temperature of the impurity problem.

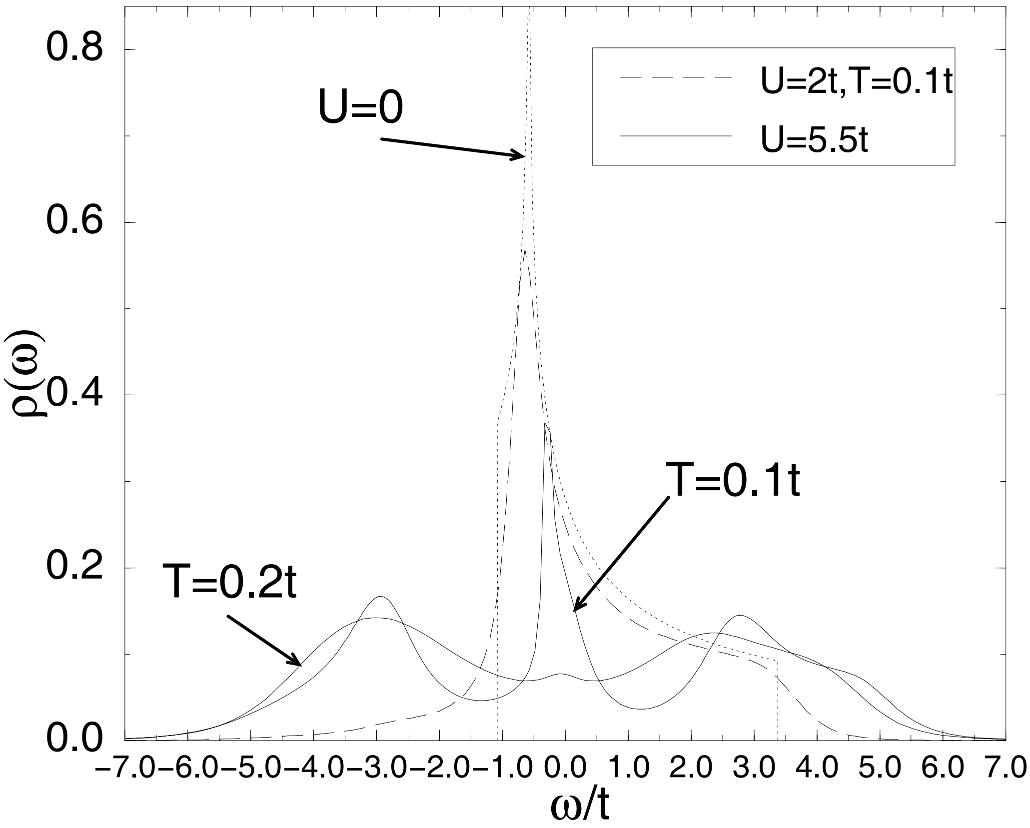
WHAT IS THE KONDO EFFECT?

A localised magnetic moment (s=1/2) interacts antiferromagnetically with conduction electrons.

- For $T < T_K$ the local moment is ”screened” by the conduction electrons and the ground state is a spin singlet.
  Impurity magnetic susceptibility $\chi_{imp}(T) \simeq \chi_0$
- For $T > T_K$ the local moment is not ”screened” $\chi_{imp}(T) \sim 1/T$

DENSITY OF STATES FROM DMFT

Triangular lattice



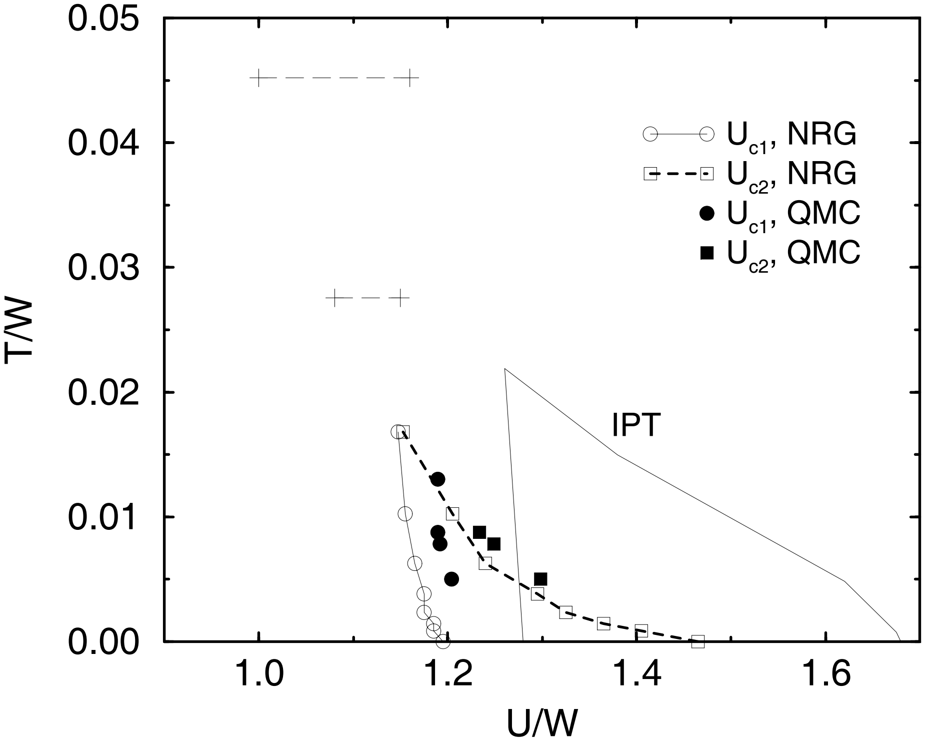
- The peak at the Fermi energy corresponds to a band of quasi-particles and only exists for $T < T_0 \sim 0.1D$.
- The broad peaks are the upper and lower Hubbard bands, separated by about U, and corresponding to incoherent excitations.

METAL-INSULATOR TRANSITION

DMFT Phase diagram of Hubbard model with significant magnetic frustration.

Bulla *et al.*, *PRB* **64**, 045103 (2001).

Temperature vs. U/t

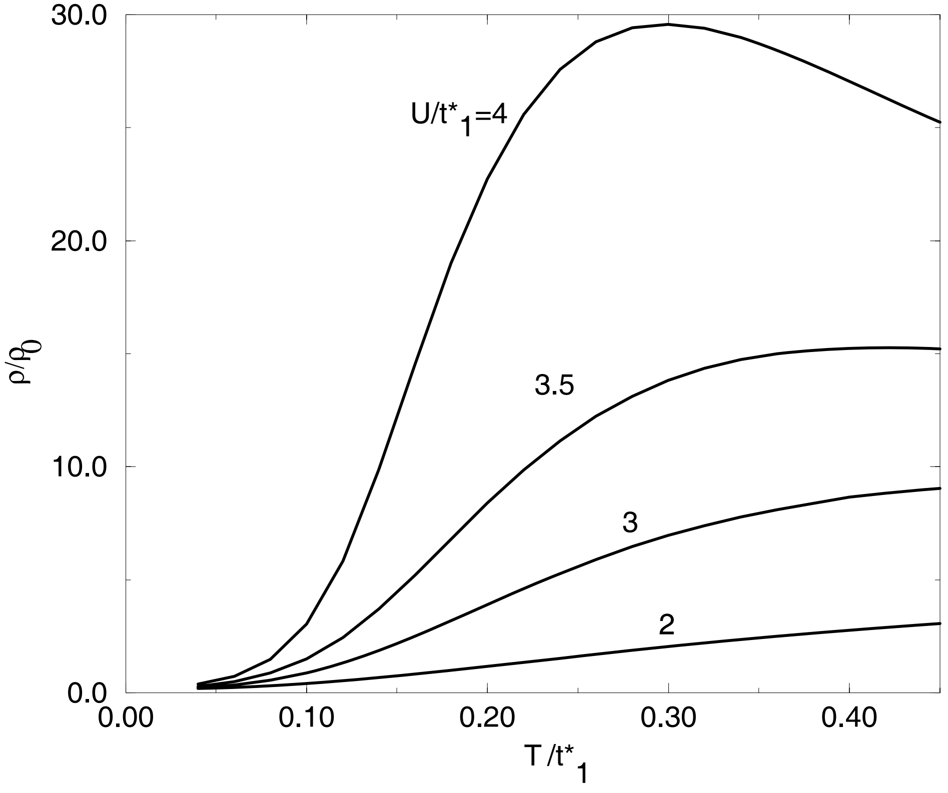


- Metal-insulator transition is first order with a critical point at $T = 0.16W$. This is as observed in κ-(BEDT-TTF)$_2$X. Limelette *et al.*, PRL **91**, 016401 (2003). Kagawa *et al.*, cond-mat/0307304.

TRANSPORT PROPERTIES FROM DMFT

Merino and RHM, *PRB* **61**, 7996 (2000).

Temperature dependence of resistivity

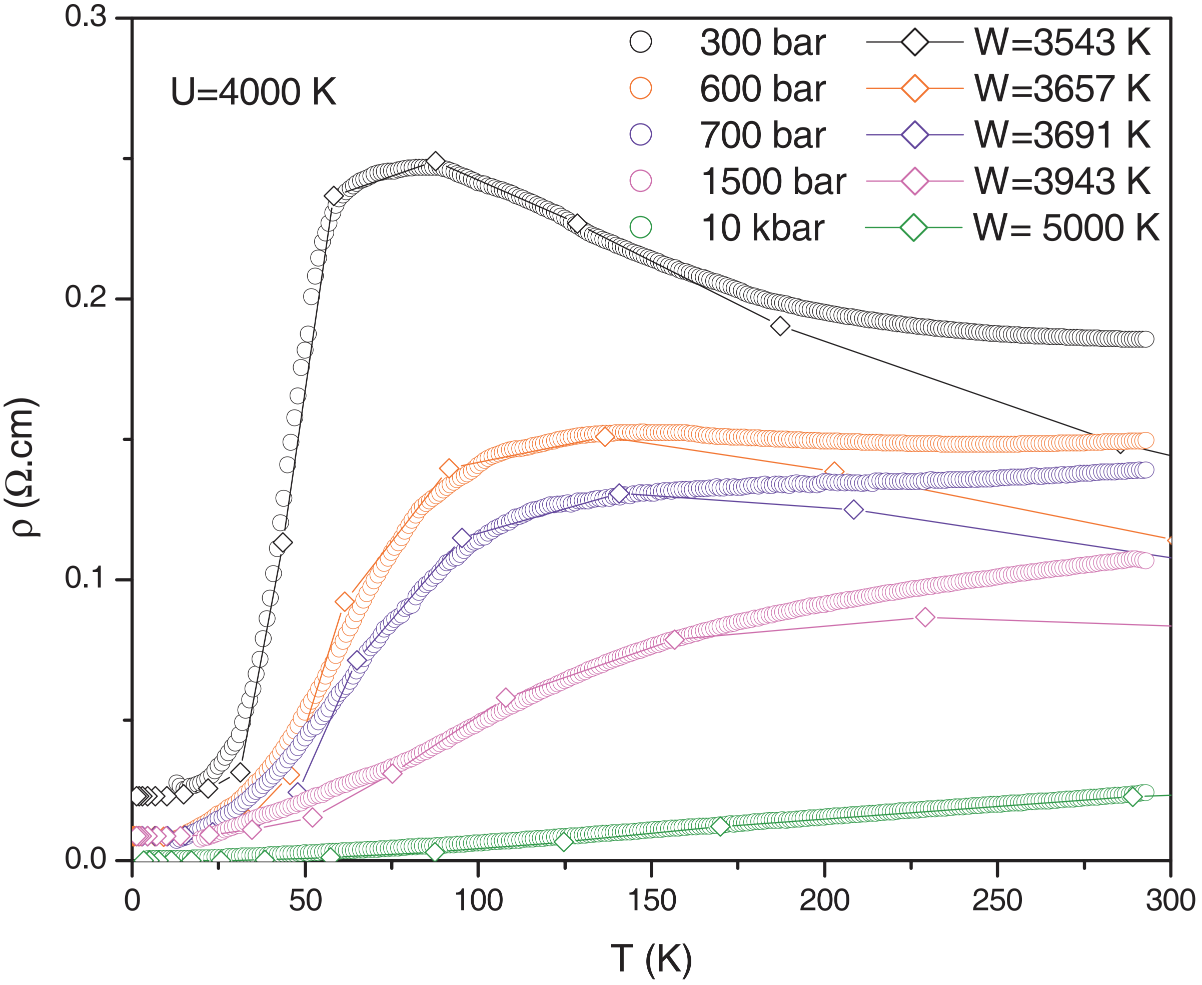


- $\rho_0 = \hbar a/e^2 \sim$ mΩcm, Mott limit.
- Resistivity $\rho(T)$ does not monotonically increase with temperature.
- Smooth crossover from $\rho \sim T^2$ at low temperatures to bad metal, (above T_0 "mean-free path" less than a lattice constant).

TRANSPORT PROPERTIES FROM DMFT

Temperature dependence of resistivity

Limelette *et al.*, PRL **91**, 016401 (2003).



TRANSPORT PROPERTIES FROM DMFT

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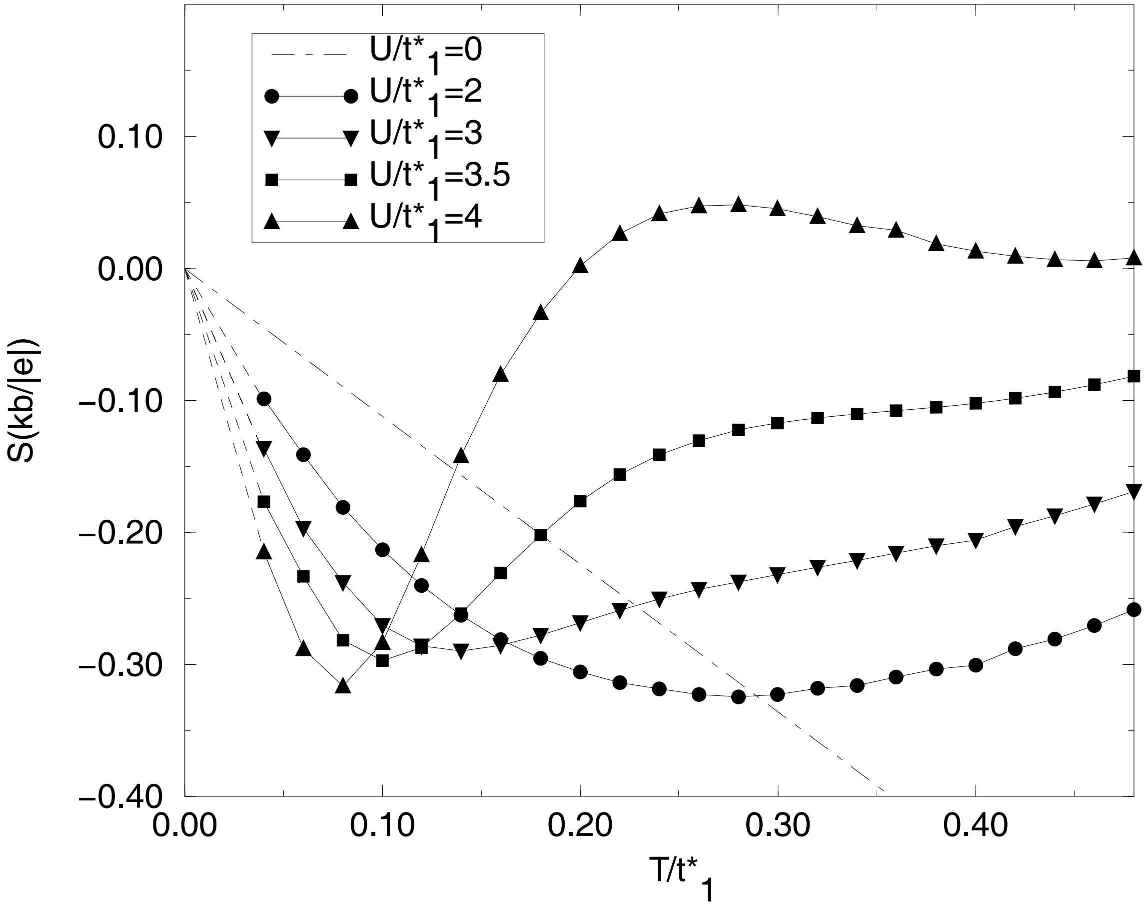
Temperature dependence of thermopower

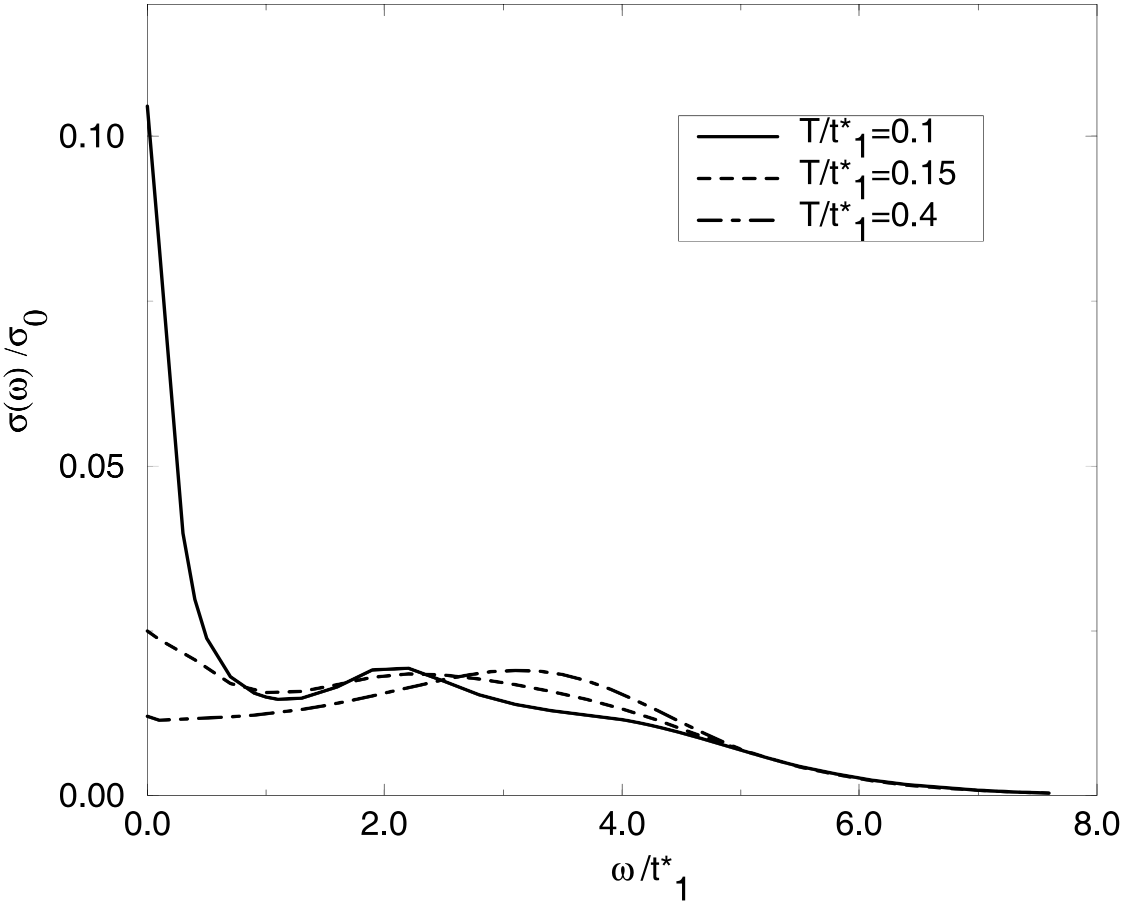


- Values of order of $k_B/e \sim 76\ \mu\mathrm{V/K}$
- Non-monotonic temperature dependence
- Feature at $T \simeq T_0$

TRANSPORT PROPERTIES FROM DMFT

Merino and RHM, *PRB* **61**, 7996 (2000).

Conductivity vs. frequency

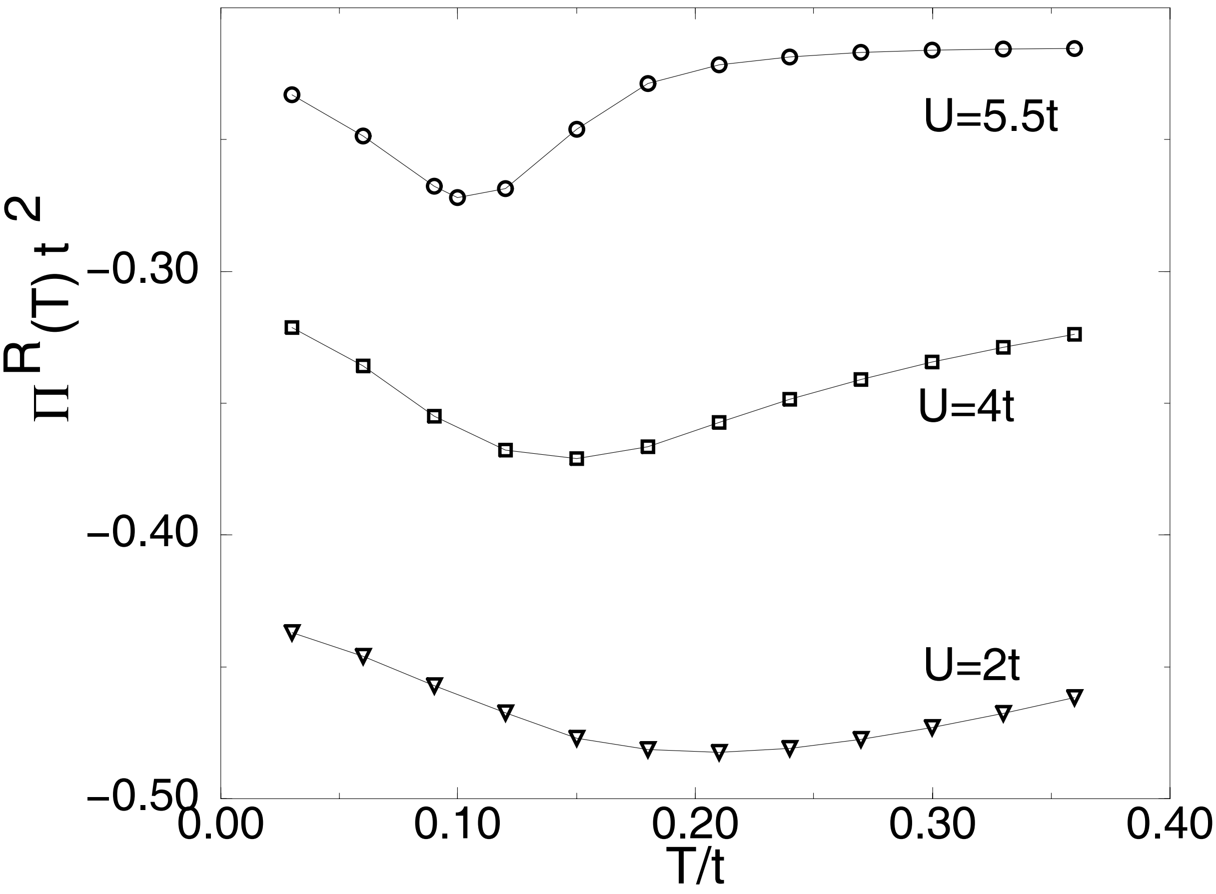


- Drude peak only occurs for $T < T_0$
- Broad feature at $\omega \sim U$ due to transitions between Hubbard bands is weakly temperature dependent.

ACOUSTIC ANOMALIES AT T_0

Merino and RHM, *PRB* **62**, 16442 (2000).

Phonon self energy is related to electronic density-density correlation function. Temperature dependence of speed of sound



- Feature at $T \simeq T_0$ grows as the Mott transition is approached.

IMPORTANT EXPERIMENTS

For $T > T_0$ the presence of local moments should be detectable with μSR.

CONCEPTUAL QUESTIONS

How does spin-charge separation enter?

In what sense are the Hubbard bands incoherent?

THEORETICAL CHALLENGES

How we allow for spatial correlations?
Cellular DMFT?
Need to describe antiferromagnetism and d-wave superconductivity.
Frequency dependence of Drude peak.

FUTURE CALCULATIONS

Magnetic susceptibility, nmr relaxation rate.

CONCLUSIONS

(1) The metallic phase of κ-(BEDT-TTF)$_2$X is a strongly correlated metal close to a Mott insulating phase. It exhibits a crossover from a Fermi liquid to a bad metal at $T_0 \sim 30$ K.

(2) The relevant theoretical model is a Hubbard model on an anisotropic triangular lattice at half-filling.

(3) Transport properties, including the coherence temperature T_0, is described by dynamical mean-field theory.

(4) Thermopower is particularly sensitive to T_0.

(5) All these ideas are equally applicable to β, β', and λ-D$_2$X.