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XII WORKSHOP ON
STRONGLY CORRELATED ELECTRON SYSTEMS

17 - 28 July 2000

A STRONGLY CORRELATED ELECTRON MODEL FOR THE QUASI-TWO-DIMENSIONAL ORGANIC SUPERCONDUCTORS KAPPA-(BEDT-TTF)2Z

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A STRONGLY CORRELATED ELECTRON MODEL FOR LAYERED ORGANIC SUPERCONDUCTORS

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Phys. Rev. B **61**, 7996 (2000).

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MAIN POINTS

(1) The layered organic superconductors κ-(BEDT-TTF)$_2$X have important similarities to the high-T_c cuprates including

- competition between antiferromagnetism and superconductivity
- unconventional metallic behaviour
- unconventional superconductivity.

(2) The relevant theoretical model is a Hubbard model on an anisotropic triangular lattice with one electron per site.

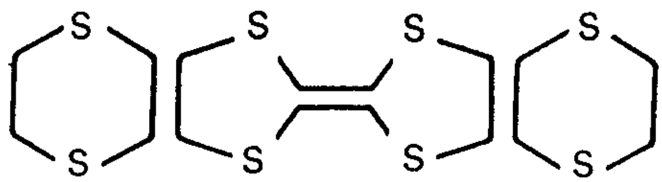
Key features are

- strongly interacting electrons
- proximity to a metal-insulator transition
- significant magnetic frustration.

R.H. McKenzie, *Comments on Condensed Matter Physics*, **18**, 309 (1998).

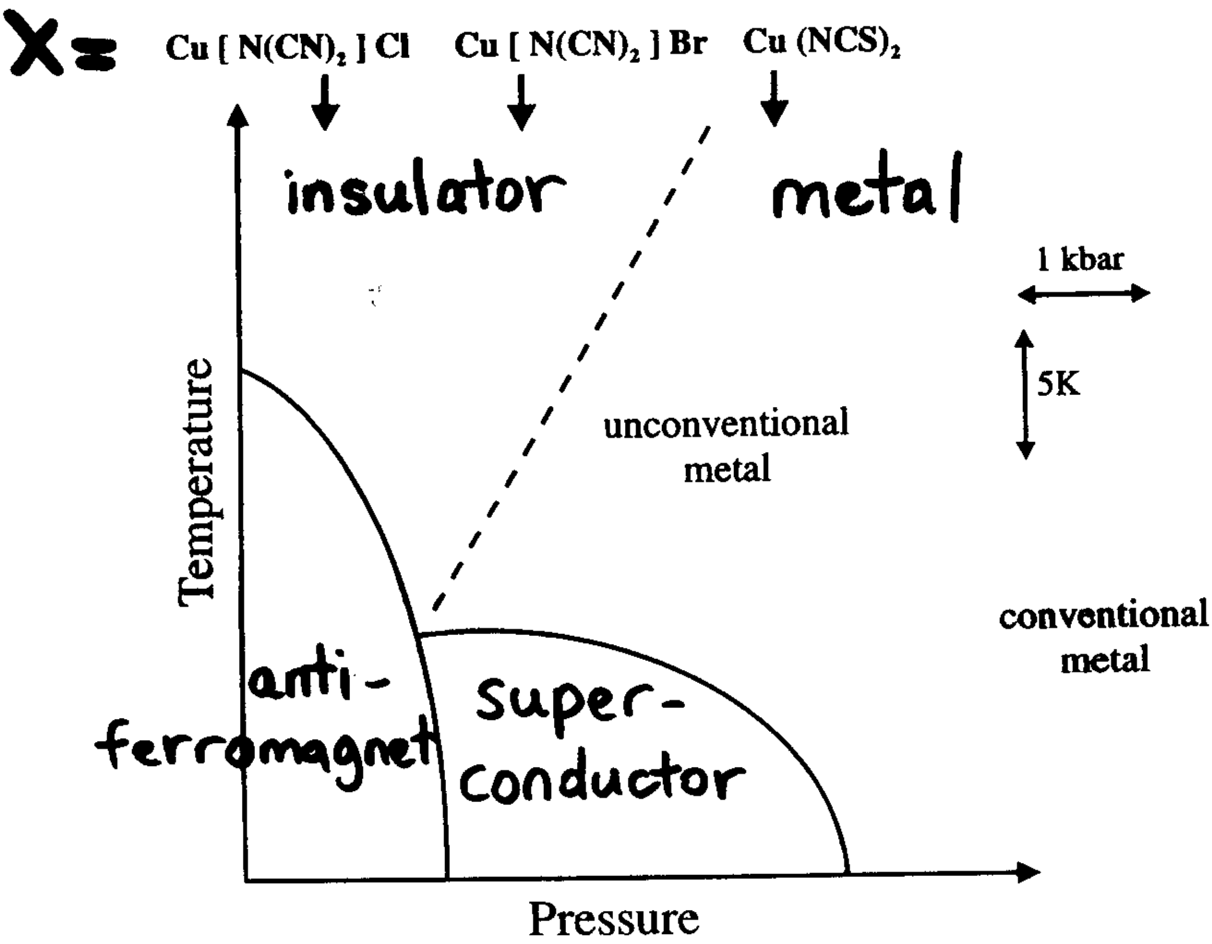
ORGANIC SUPERCONDUCTORS

- Bechgaard salts - $(TMTSF)_2X$ ($X=ClO_4, PF_6, ..$) Quasi-one-dimensional electronic properties
- Fullerenes - M_3C_{60}
- BEDT-TTF salts - e.g., α-$(BEDT\text{-}TTF)_2X$



Conducting layers of BEDT-TTF molecules are separated by layers of anions X.
Quasi-two-dimensional electronic properties
Greek letters α, β, κ, and θ denote the stacking pattern of the BEDT-TTF molecules in each layer.

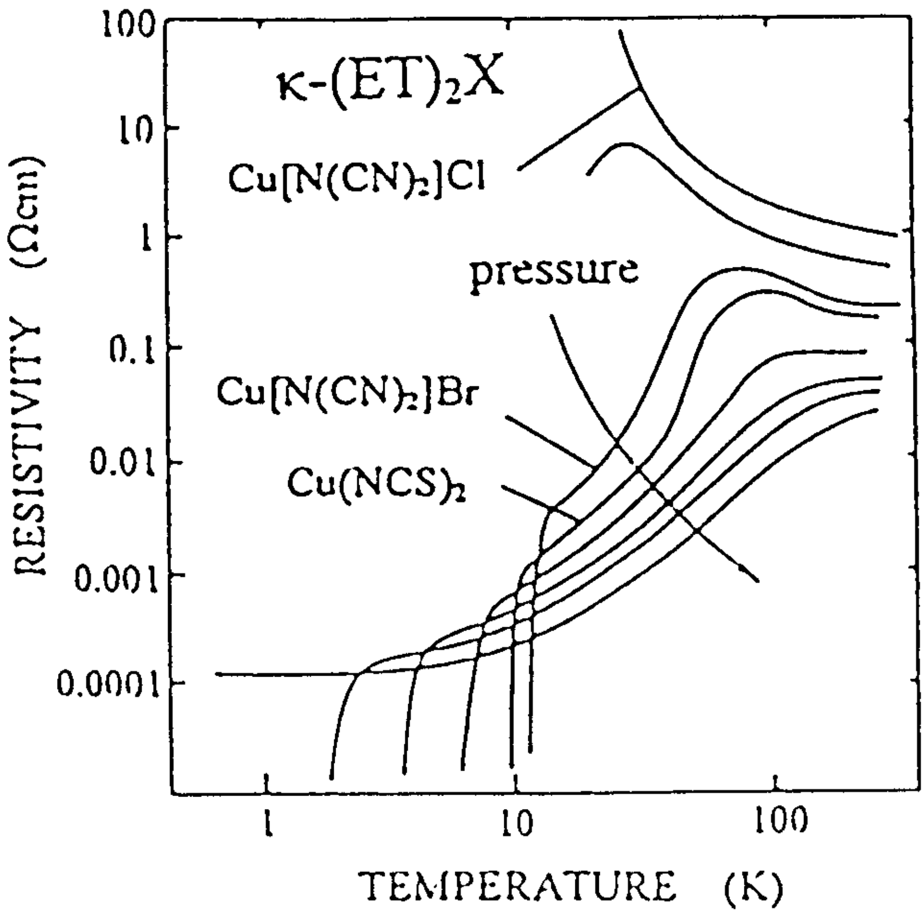
PHASE DIAGRAM FOR LAYERED MOLECULAR CRYSTALS κ-(BEDT-TTF)$_2$X



- unconventional metal near metal-insulator transition
- competition between superconductivity and antiferromagnetism
- similar to high-T_c cuprates except pressure plays the role of doping

UNCONVENTIONAL METALLIC PROPERTIES

Temperature dependence of resistivity

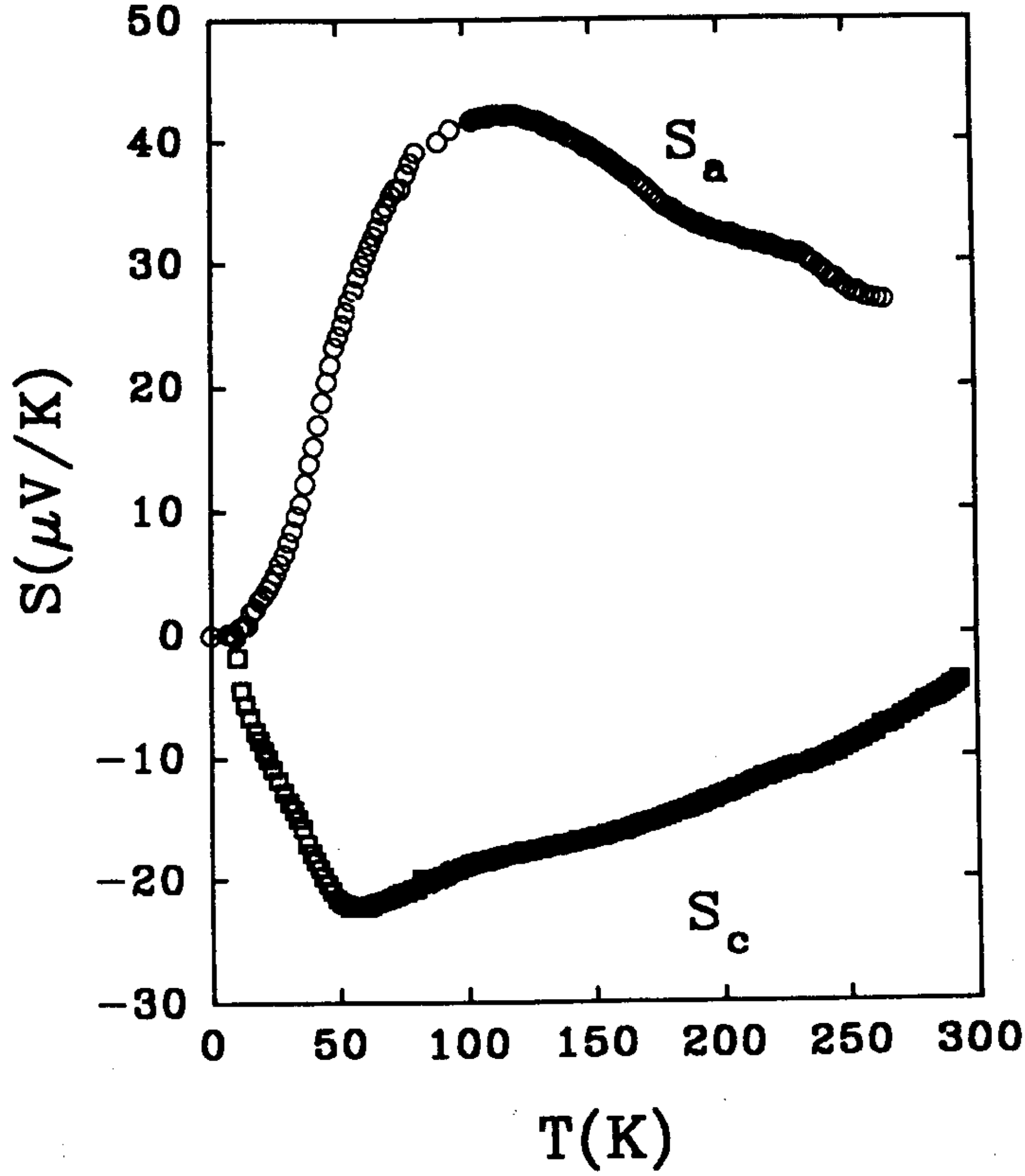


- A low temperatures, $\rho \sim AT^2$ at low T, suggesting a Fermi liquid.
- At high temperatures, $\rho > \hbar a/e^2$ ('bad metal') so the mean-free path is less than a lattice constant.
- Non-monotonic temperature dependence

UNCONVENTIONAL METALLIC PROPERTIES

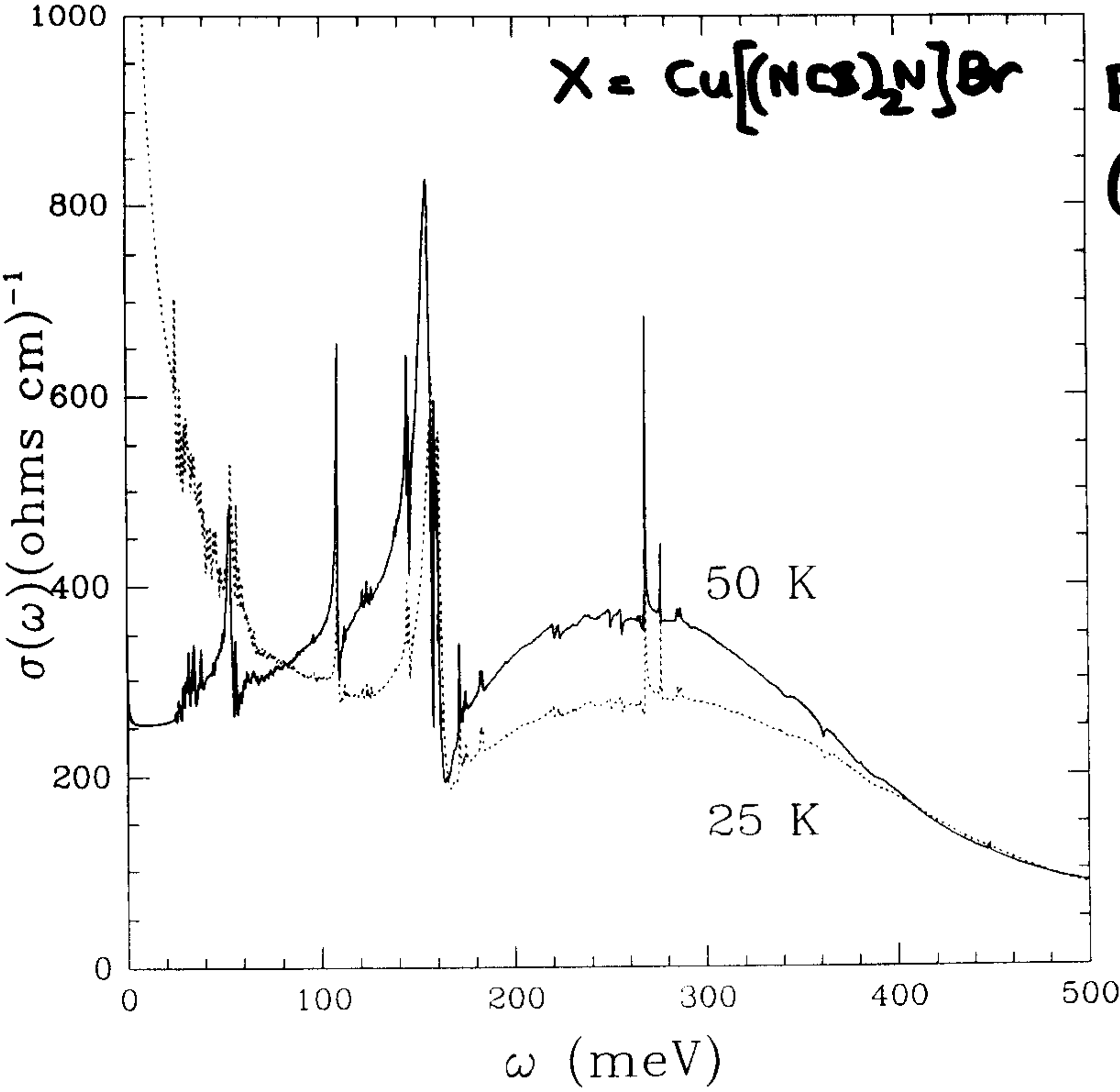
Temperature dependence of thermopower

κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br



- Values of the order of k_B/e.
- Non-monotonic temperature dependence

UNCONVENTIONAL METALLIC PROPERTIES
Temperature dependence of optical conductivity



Eldridge (UBC)

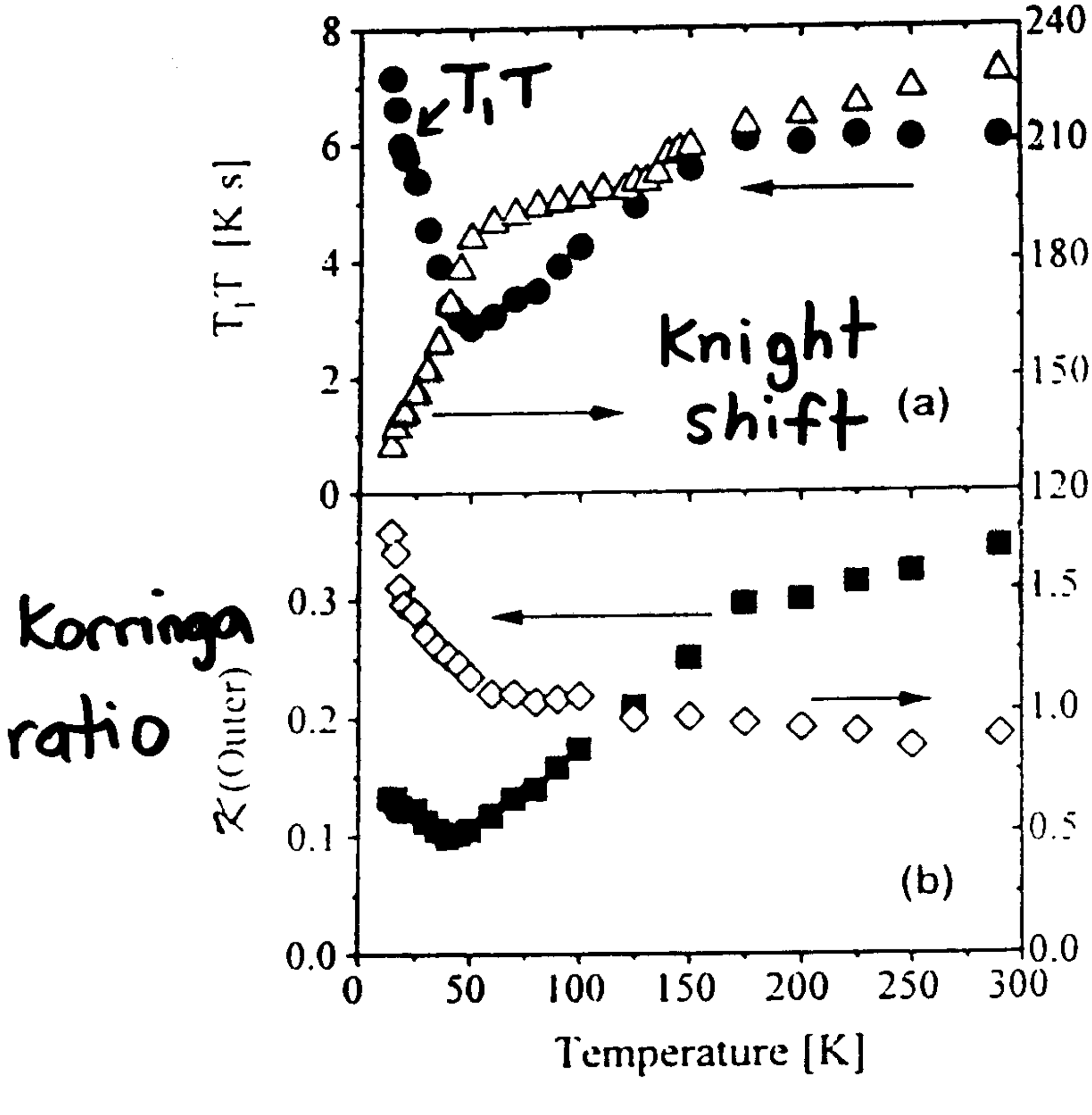
- No Drude ($\omega = 0$) peak above 50 K
- Broad peak around 300 meV.

Is only a "conventional" metal below $T_0 \sim 50K \ll$ Fermi energy E_F

[illegible]

TEMPERATURE DEPENDENCE OF NMR

κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br



UNCONVENTIONAL SUPERCONDUCTIVITY

Evidence for *gapless excitations* or nodes in the gap is that some properties are not exponentially activated but have a power law dependence on temperature.

- nmr relaxation rate
  - $1/T_1 \sim T^3$
  - no Hebel-Slichter peak
- Thermal conductivity $\kappa \sim T$
- specific heat (controversial)
- penetration depth (controversial)
- magneto-optical (controversial)

Evidence for *singlet pairing*
The nmr Knight shift $K(T) \to 0$ as $T \to 0$.
The upper critical field B_{c_2} for the field in the layers is comparable to the Pauli paramagnetic (Clogston) limit at which the Zeeman splitting destroys singlets.

SUPERFLUID DENSITY VERSUS TEMPERATURE

Carrington et al., PRL 83, 4172 (1999)

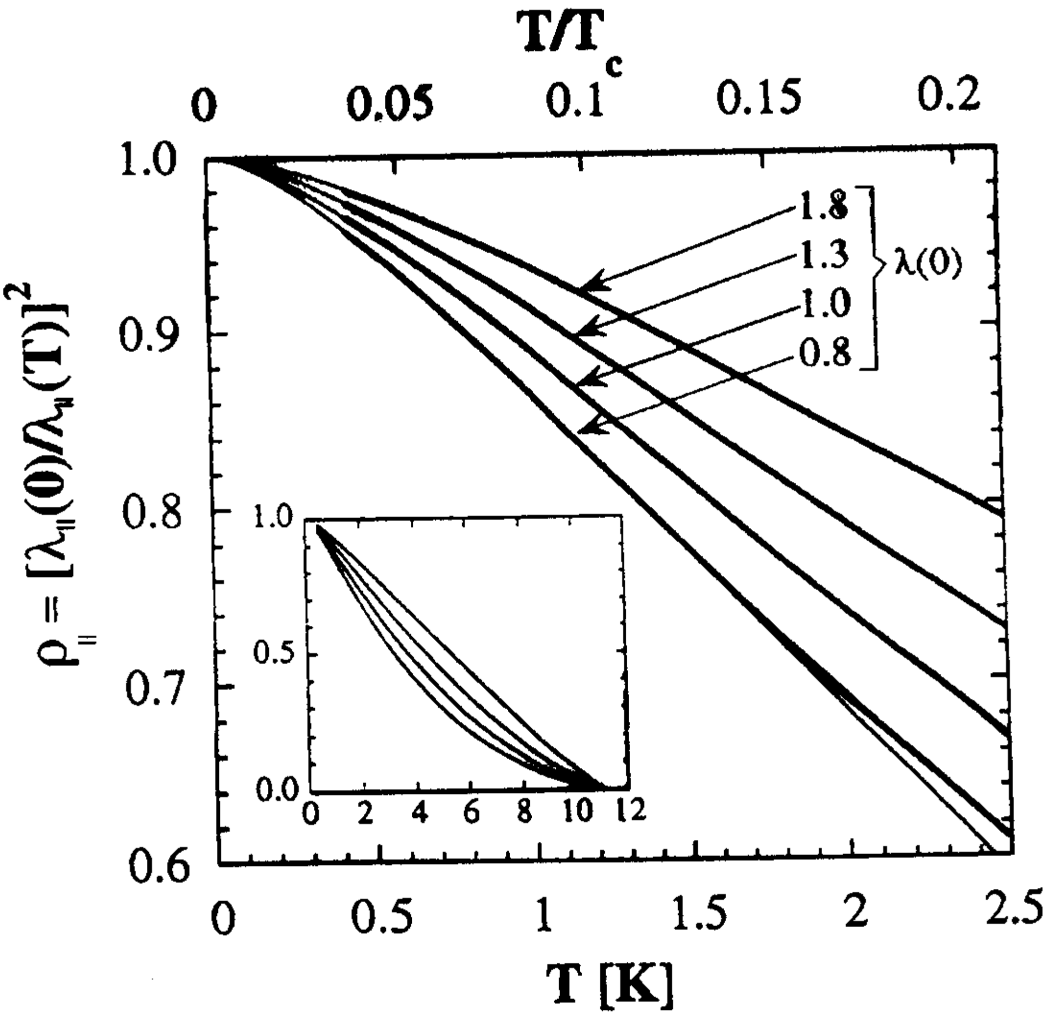
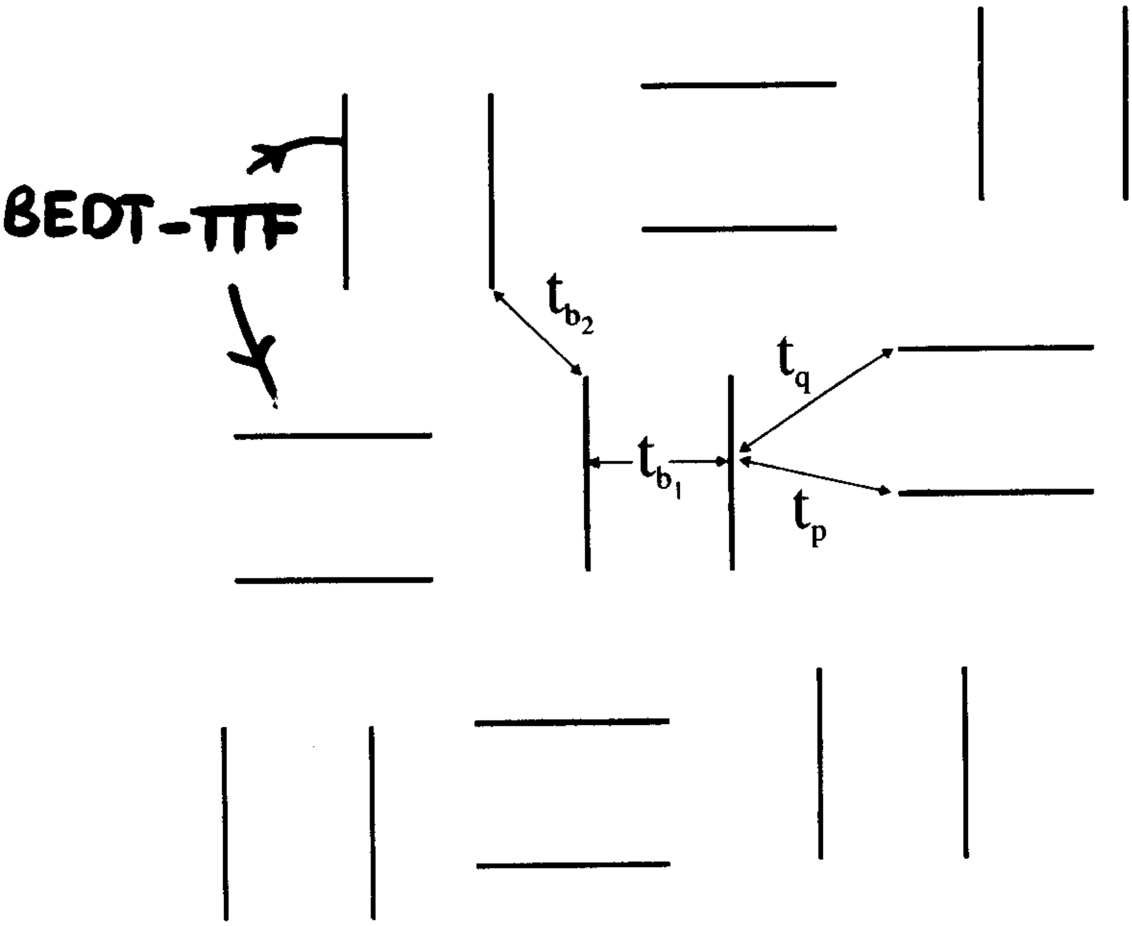


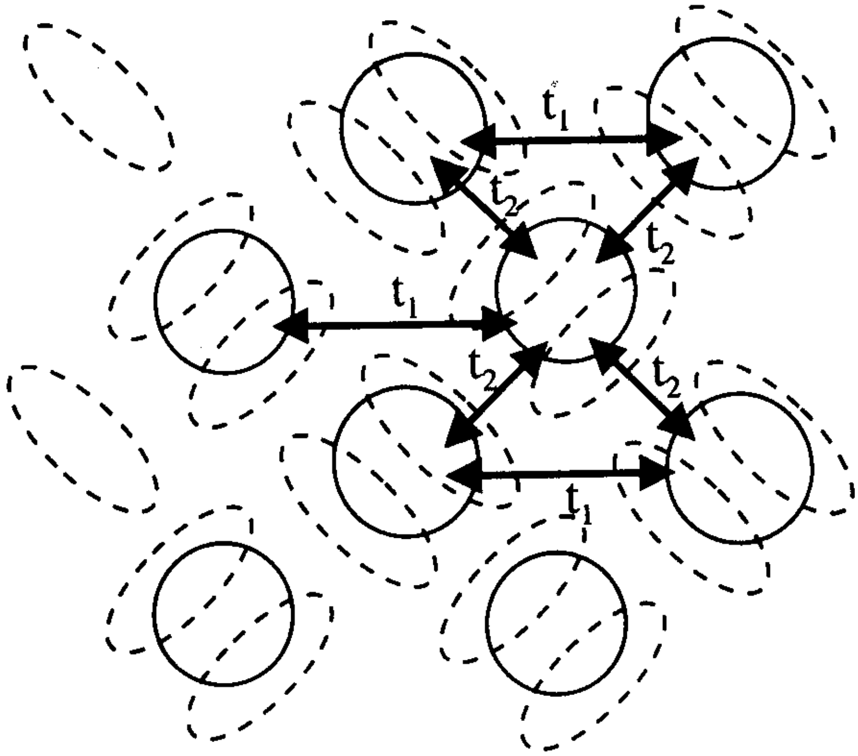
FIG. 2. In-plane superfluid density $\rho_{\parallel} = \lambda_{\parallel}^2(0)/\lambda_{\parallel}(T)^2$ calculated from the $\Delta\lambda_{\parallel}(T)$ data in Fig. 1, for several values of $\lambda_{\parallel}(0)$. The thin lines are fits to the data with Eq. (1). The inset shows the same data over a wider temperature range.

DIMER STRUCTURE OF κ-(BEDT-TTF)$_2$X



Hopping parameters between neighbouring BEDT-TTF molecules within each layer. The bonding and anti-bonding orbitals on each dimer are split by $2t_{b_1} \gg t_{b_2}, t_p, t_q$. Thus, the inter-dimer hopping can be viewed as a perturbation.

DIMER MODEL (Kino and Fukuyama)



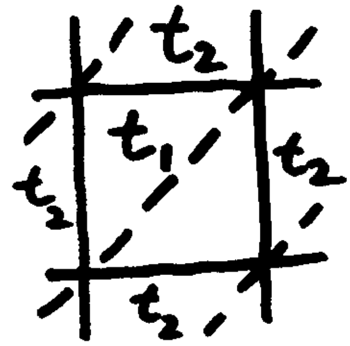
There is one hole per dimer.
The dimers form an *anisotropic triangular lattice* with different hopping in the horizontal (t_1) and slanted directions (t_2).
U is the Coulomb repulsion between two holes on a dimer. From quantum chemistry U is larger than the band width.
This is a strongly correlated system.
Magnetic frustration results from competition between t_1 and t_2.

THE RELEVANT HUBBARD MODEL

$$H = -t_1 \sum_{<\mathbf{ij}>,\sigma} (c^\dagger_{\mathbf{i},\sigma} c_{\mathbf{j},\sigma} + h.c.)$$

$$-t_2 \sum_{<\mathbf{in}>,\sigma} (c^\dagger_{\mathbf{i},\sigma} c_{\mathbf{n},\sigma} + h.c.)$$

$$+U \sum_{\mathbf{i}} (n_{\mathbf{i}\uparrow} - \frac{1}{2})(n_{\mathbf{i}\downarrow} - \frac{1}{2}) + \mu \sum_{\mathbf{i},\sigma} n_{\mathbf{i}\sigma}$$



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

Insight can be obtained by considering limits for which there are known results.

$t_1 = 0$ square lattice

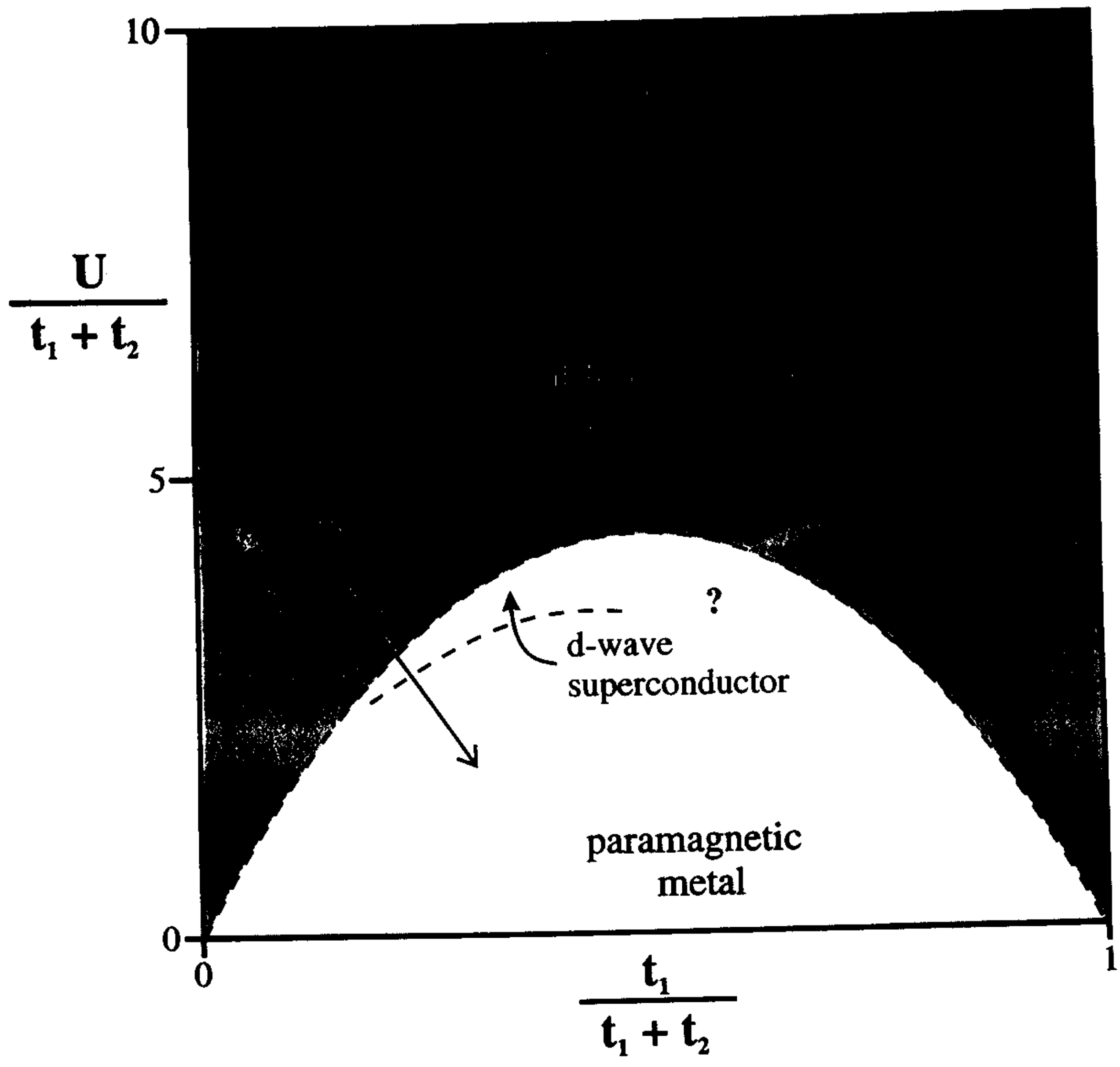
$t_1 = t_2$ triangular lattice

$t_2 = 0$ decoupled chains

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.

TENTATIVE PHASE DIAGRAM FOR THE HUBBARD MODEL ON AN ANISOTROPIC TRIANGULAR LATTICE

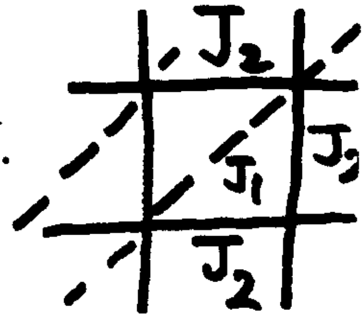
Zero temperature



MAGNETIC ORDER IN THE INSULATING PHASE

For $U \gg t_1, t_2$ the spin degrees of freedom are described by the Heisenberg model

$$H = J_1 \sum_{\langle \mathbf{ij} \rangle} \vec{S}_{\mathbf{i}} \cdot \vec{S}_{\mathbf{j}} + J_2 \sum_{\langle \mathbf{in} \rangle} \vec{S}_{\mathbf{i}} \cdot \vec{S}_{\mathbf{n}}$$



where $J_1 = 4t_1^2/U$ and $J_2 = 4t_2^2/U$.

The classical ground state is

$$< \vec{S}_{\vec{r}} > \sim \exp(i\vec{r} \cdot \vec{Q})$$

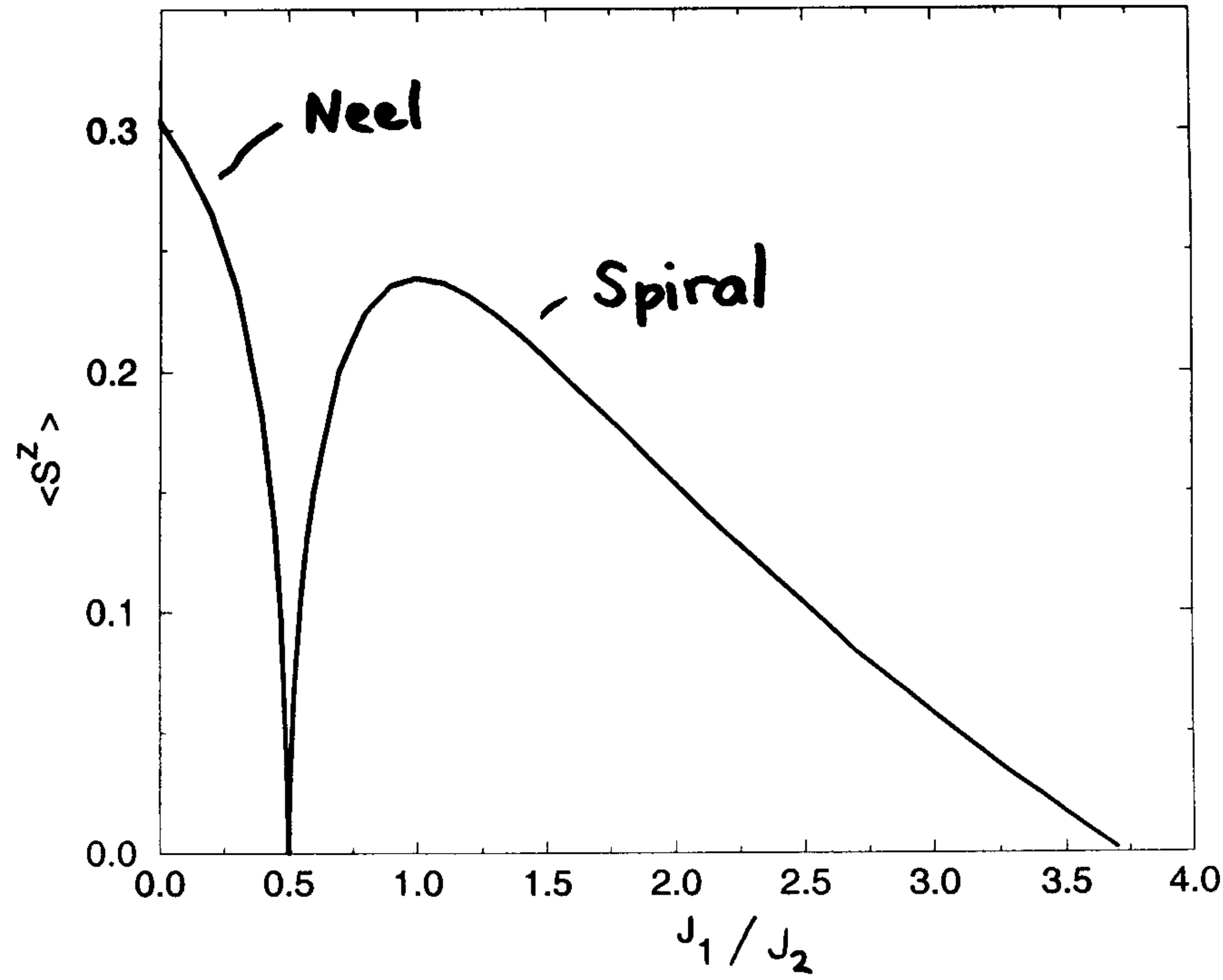
where $\vec{Q} = (Q, Q)$
Collinear Neel order, $Q = \pi$ for $J_1 < J_2/2$.
Non-collinear order, $Q = \arccos(-J_2/2J_1)$ for $J_1 > J_2/2$.

What is the effect of quantum fluctuations?

LINEAR SPIN-WAVE THEORY

Merino *et al.*, cond-mat/9812429 J. Phys.: CM 11, 2965 (1999)

Magnetization of the spiral state



Ground state may be quantum disordered near $J_1 = J_2/2$ and for $J_1 > 4J_2$.

UNCONVENTIONAL SUPERCONDUCTIVITY

Is there superconductivity in the model?

Yes. According to Quantum Monte Carlo calculations and the random-phase approximation (RPA) and the fluctuation-exchange approximation (FLEX). It is mediated by spin fluctuations.

What is the relationship between the magnetic ground state of the insulating phase and the symmetry of Cooper pairs in the superconducting phase?

Neel (π, π) order leads to d-wave singlet superconductivity, as in the cuprates.

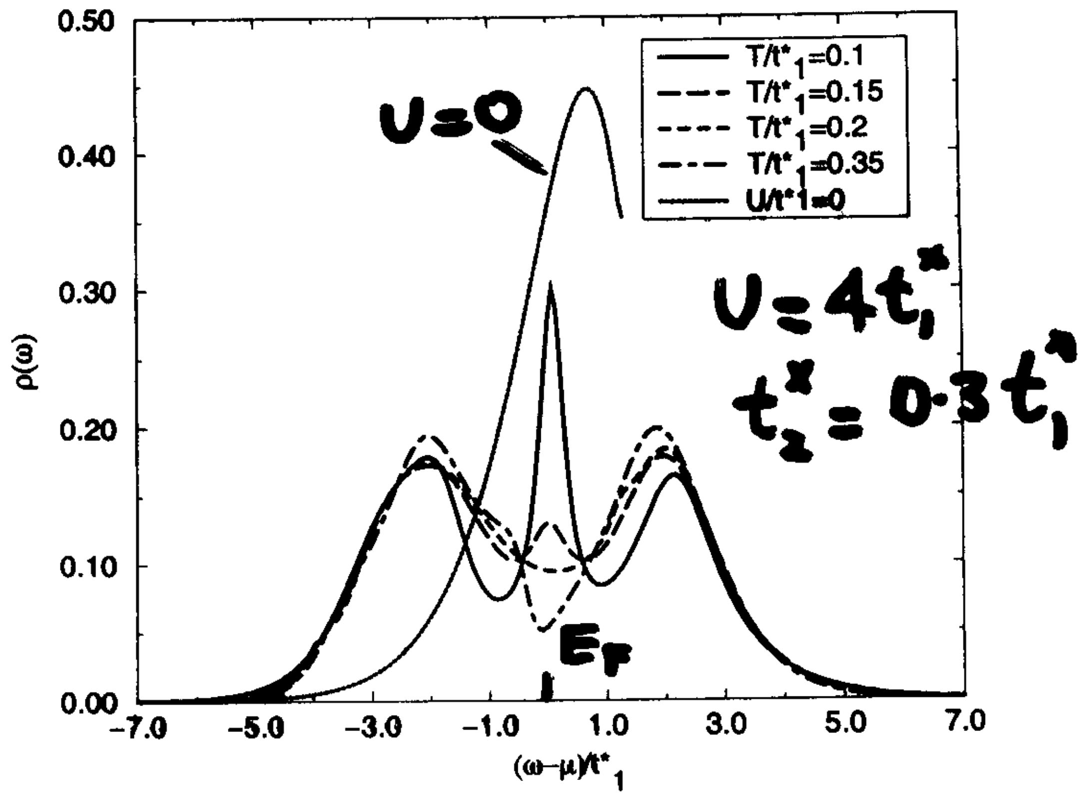
The 120 degree spiral order of the isotropic triangular lattice leads to Cooper pairing in the s-wave triplet odd-frequency channel.

M. Votja and E. Dagotto, PRB, 1999; J. Schmalian, PRL **81**, 4232 (1998); H. Kino and H. Kontani, cond-mat/9807147; H. Kondo and T. Moriya, cond-mat/980732 K. Kuroki and H. Aoki, cond-mat/9812026.

DYNAMICAL MEAN-FIELD THEORY

Georges et al. Rev. Mod. Phys. 68, 13 (1996)

Spectral density



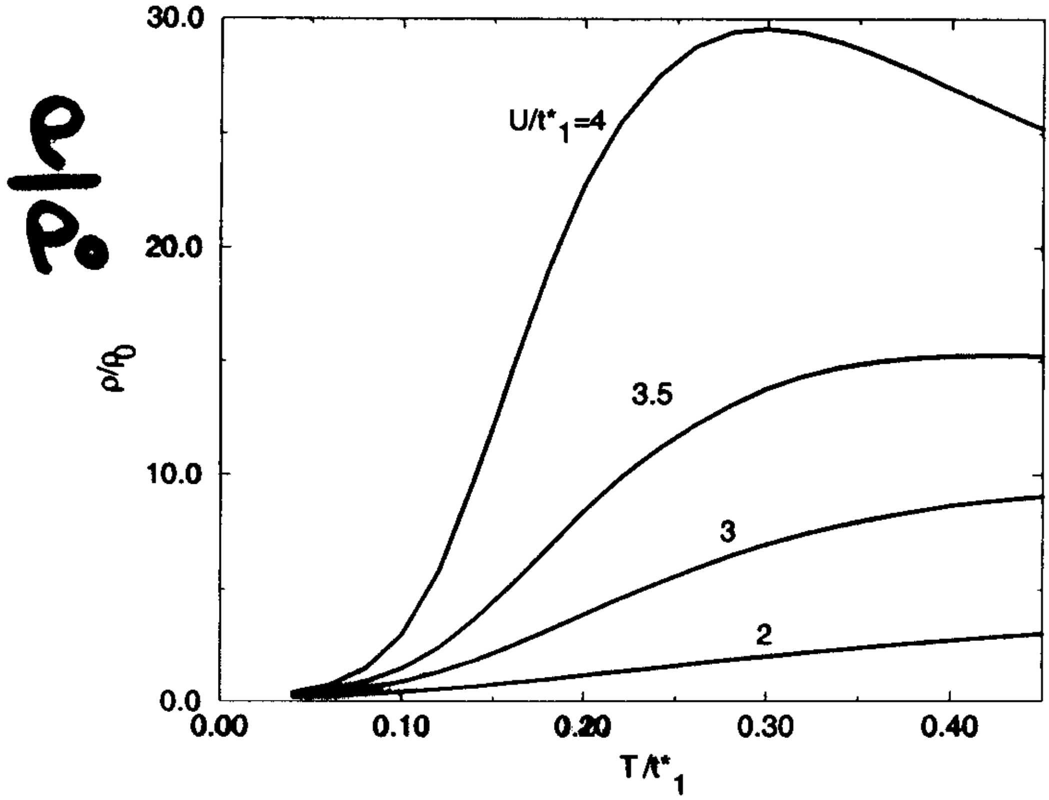
• Low energy scale given by T_0, analogue of the Kondo temperature.

• Crossover from coherent Fermi liquid excitations for $T \ll T_0$ to incoherent excitations at $T > T_0$. This gives the strong temperature dependence of transport quantities.

Merino and McKenzie, cond-mat/990904

CROSSOVER TO INCOHERENT EXCITATIONS

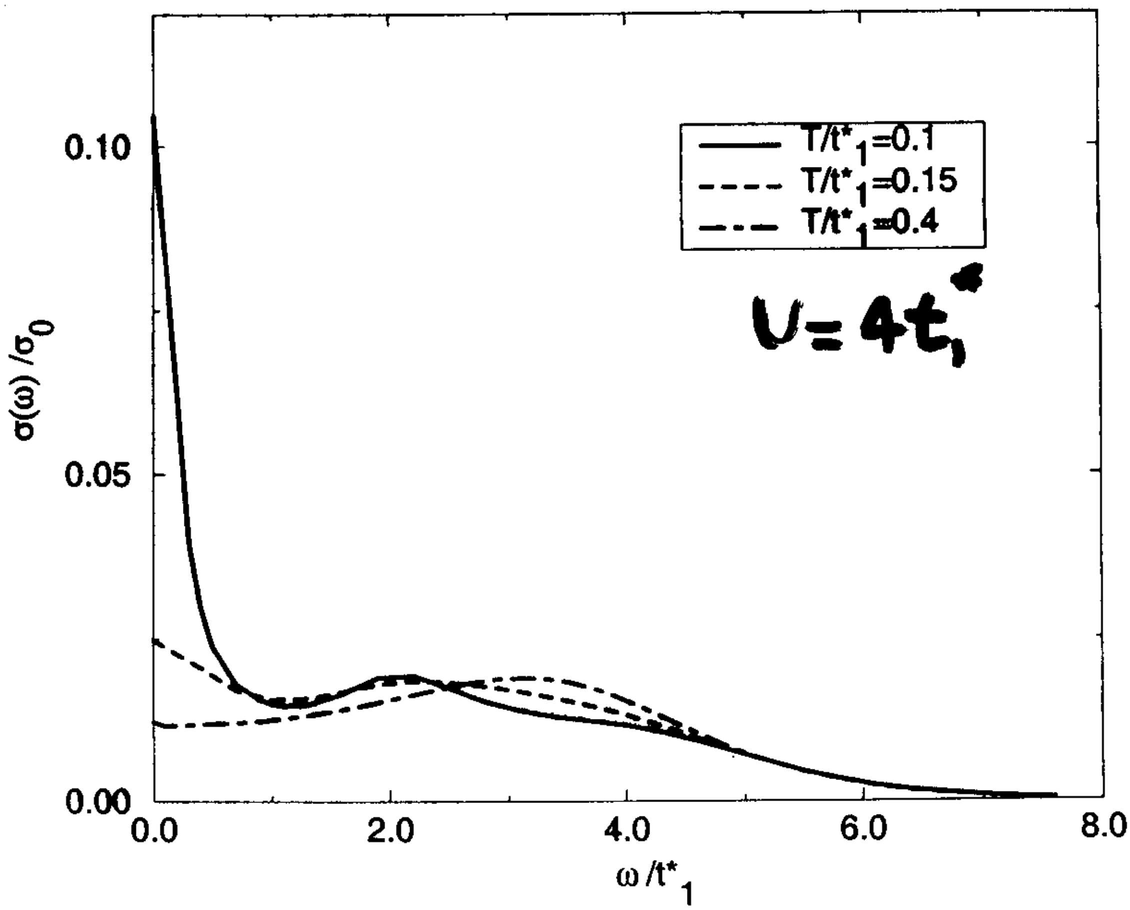
Temperature dependence of resistivity



- $\rho_0 = \hbar a/e^2 \sim m\Omega$cm, Mott limit
- For mean-free path smaller than the lattice parameter, the resistivity continues to increase.
- Crossover from T^2 Fermi liquid behaviour to a bad metal.

OPTICAL CONDUCTIVITY

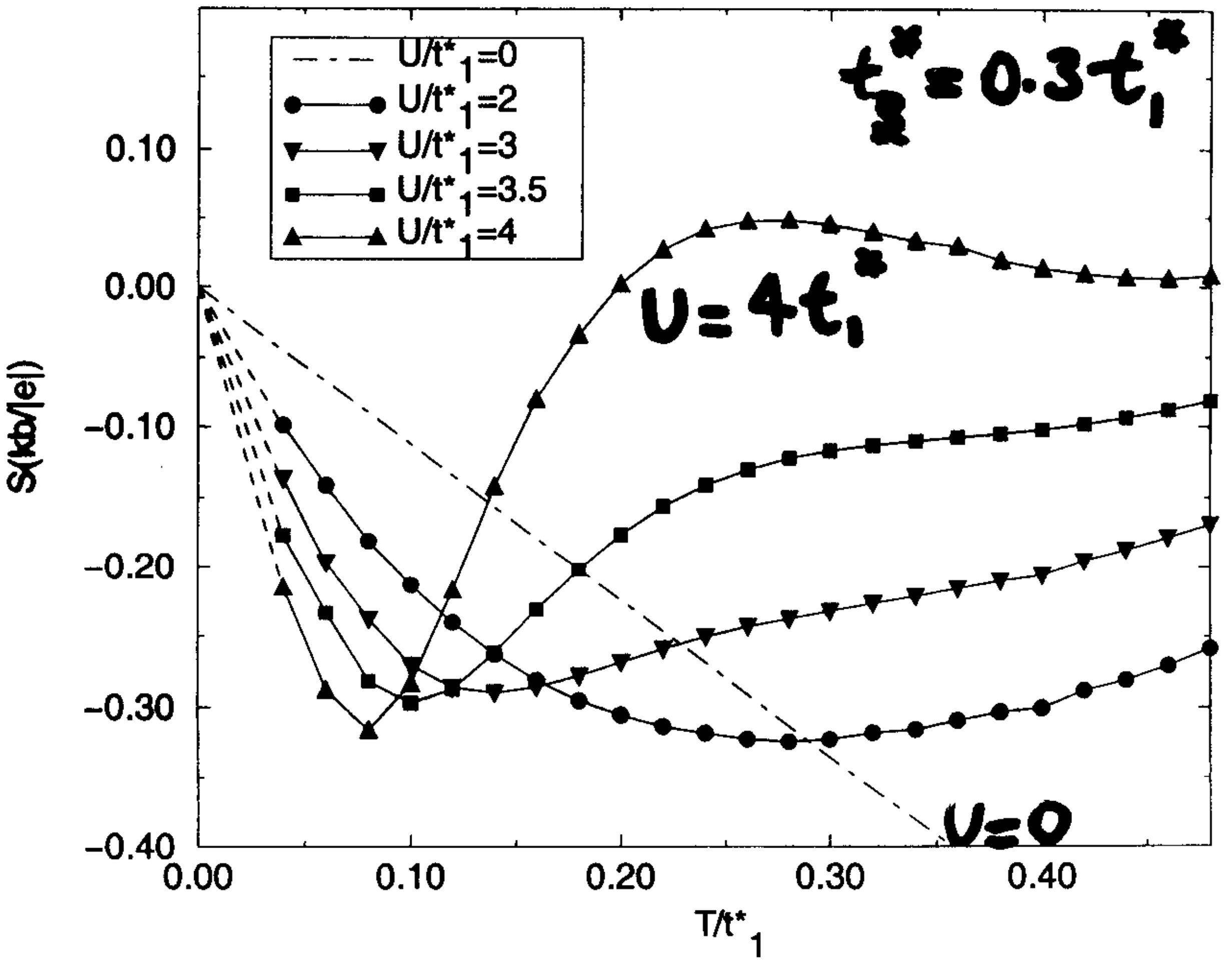
Frequency-dependence of conductivity



- The Drude peak only appears for temperatures of the order of T_0 or lower.
- Broad peak at $\omega \sim U = 4t_1^*$ is due to transitions from the lower to the upper Hubbard band.

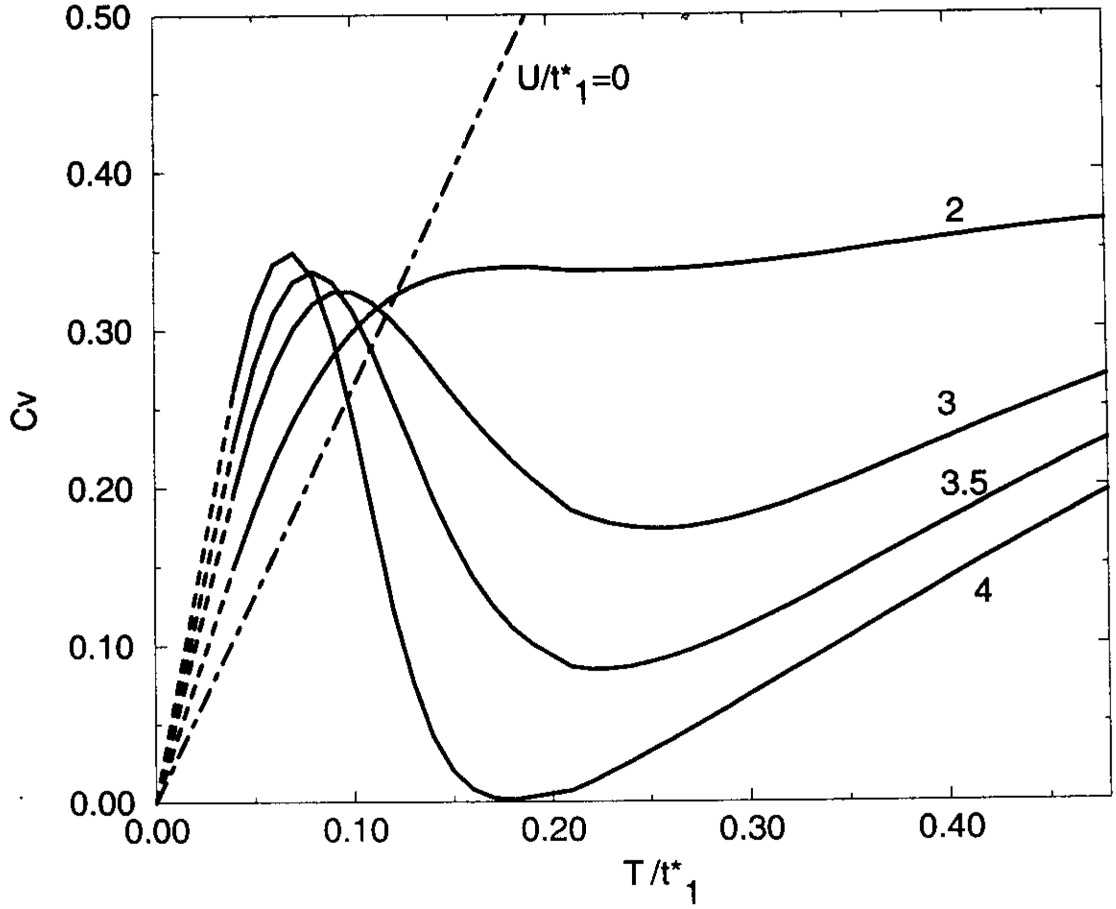
CROSSOVER TO INCOHERENT EXCITATIONS

Temperature dependence of thermopower



- Minimum associated with Kondo temperature and loss of coherence.
- Values of order $S \sim k_B/e$

Temperature dependence of specific heat



- Peak associated with Kondo temperature and loss of coherence.
- In the thermopower the peak (minimum) will *not* be masked by the phonon contribution.

CONCLUSIONS

(1) The layered organic superconductors κ-(BEDT-TTF)$_2$X have important similarities to the high-T_c cuprates including

- competition between antiferromagnetism and superconductivity
- unconventional metallic behaviour
- unconventional superconductivity.

(2) The relevant theoretical model is a Hubbard model on an anisotropic triangular lattice. Key features are

- strong correlations
- proximity to a metal-insulator transition
- significant magnetic frustration.

Insights can be gained from dynamical mean-field theory.

Detailed theoretical studies should be made of this model. Can it produce superconductivity?

OPEN QUESTIONS

Theory

- Is there superconductivity in the model?
- Are inhomogeneous metallic (stripe) phases possible?
- What are the simplest microscopic models for α, β'' and θ?

Postdoc available

Experiment

- Is there a 'smoking gun' experimental test of unconventional superconductivity?
- Systematically study the metallic phase as the metal-insulator transition is approached.
- Quantitative measures of spin fluctuations (neutron scattering).
- Thermopower is a sensitive probe of the destruction of Fermi liquid quasiparticles.