Quantum spin liquids and the Mott transition

T. Senthil (MIT)

D. Mross and T. Senthil, arxiv, ’10;

T. Grover, N. Trivedi, T. Senthil, P.A. Lee, PR B 10.

T. Senthil, PR B 08

D. Podolsky, A. Paramekanti, Y.B. Kim, T. Senthil, PRL 09

T. Senthil, P.A. Lee, PRL 09

S.S. Lee, P.A. Lee, T. Senthil, PRL 06

States of quantum magnetism

Ferromagnetism: May be 600 BC

$$|\uparrow\uparrow\uparrow\uparrow \ldots\ldots\ldots\rangle$$

Antiferromagnetism: 1930s

$$|\uparrow\downarrow\uparrow\downarrow \ldots\ldots\ldots\rangle$$

Key concept of broken symmetry.

Prototypical ground state wavefunction:
direct product of local degrees of freedom

Short range quantum entanglement.

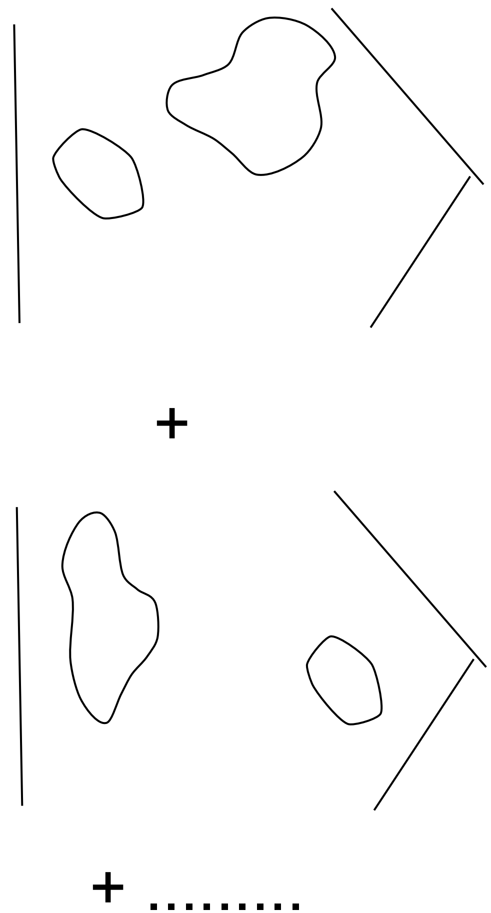
1930s- present: elaboration of broken symmetry and other
states with short range entanglement

Last ≈ 10 years

Experimental discovery of quantum spin liquid state*.

Qualitatively new kind of state of matter.
Long range quantum entanglement: Prototypical ground state wavefunction

Not a direct product of local degrees of freedom.



Many new phenomena - emergence of fractional quantum numbers.

New conceptual and technical theoretical tools to understand.

May be also new kinds of experimental probes will be most useful.

* In d > 1

What is a quantum spin liquid?

Rough description: Quantum paramagnet that does not break any symmetries of microscopic Hamiltonian.

More precise: Mott insulator with ground state <u>NOT</u> smoothly connected to band insulator.

Well known in $d=1$ spin chains

Some natural questions

Can quantum spin liquids exist in d > 1? (Anderson '73)

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Theoretical question

Do quantum spin liquids exist in d > 1?

Experimental question

Some natural questions

Can quantum spin liquids exist in d > 1? (Anderson '73)

Theoretical question: YES!! (work of many people in last 20 years)

Do quantum spin liquids exist in d > 1?

Experimental question: Remarkable new materials possibly in spin liquid phases

Organics κ-$(ET)_2 Cu(CN_3)_2$; Kagome $ZnCu(OH)_2Cl_2$,

$EtMe_3Sb[Pd(dmit)_2]_2$; Hyper Kagome $Na_4Ir_3O_8$

2d solid He-3

Where might we find quantum spin liquids?

- Geometrically frustrated quantum magnets

- ``Intermediate'' correlation regime

Eg: Mott insulators that are not too deeply into the insulating regime

Where might we find quantum spin liquids?

- Geometrically frustrated quantum magnets

  Kagome magnets ?

- ``Intermediate'' correlation regime

Eg: Mott insulators that are not too deeply into the insulating regime

Perhaps more promising in experiments ?

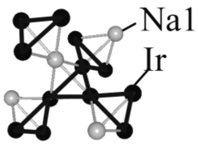
Some candidate materials

$\kappa - (ET)_2Cu_2(CN)_3$

$EtMe_3Sb[Pd(dmit)_2]_2$

Quasi-2d, approximately isotropic triangular lattice; best studied candidate spin liquids

$Na_4Ir_3O_8$



Three dimensional `hyperkagome' lattice

Close to pressure driven Mott transition: `weak' Mott insulators

$ZnCu_3(OH)_6Cl_2$

Volborthtite, ...

2d Kagome lattice (`strong' Mott insulator)

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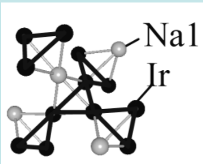

Three dimensional `hyperkagome' lattice

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Volborthtite, ...

2d Kagome lattice (`strong' Mott insulator)

Some phenomena in experiments

ALL candidate materials:

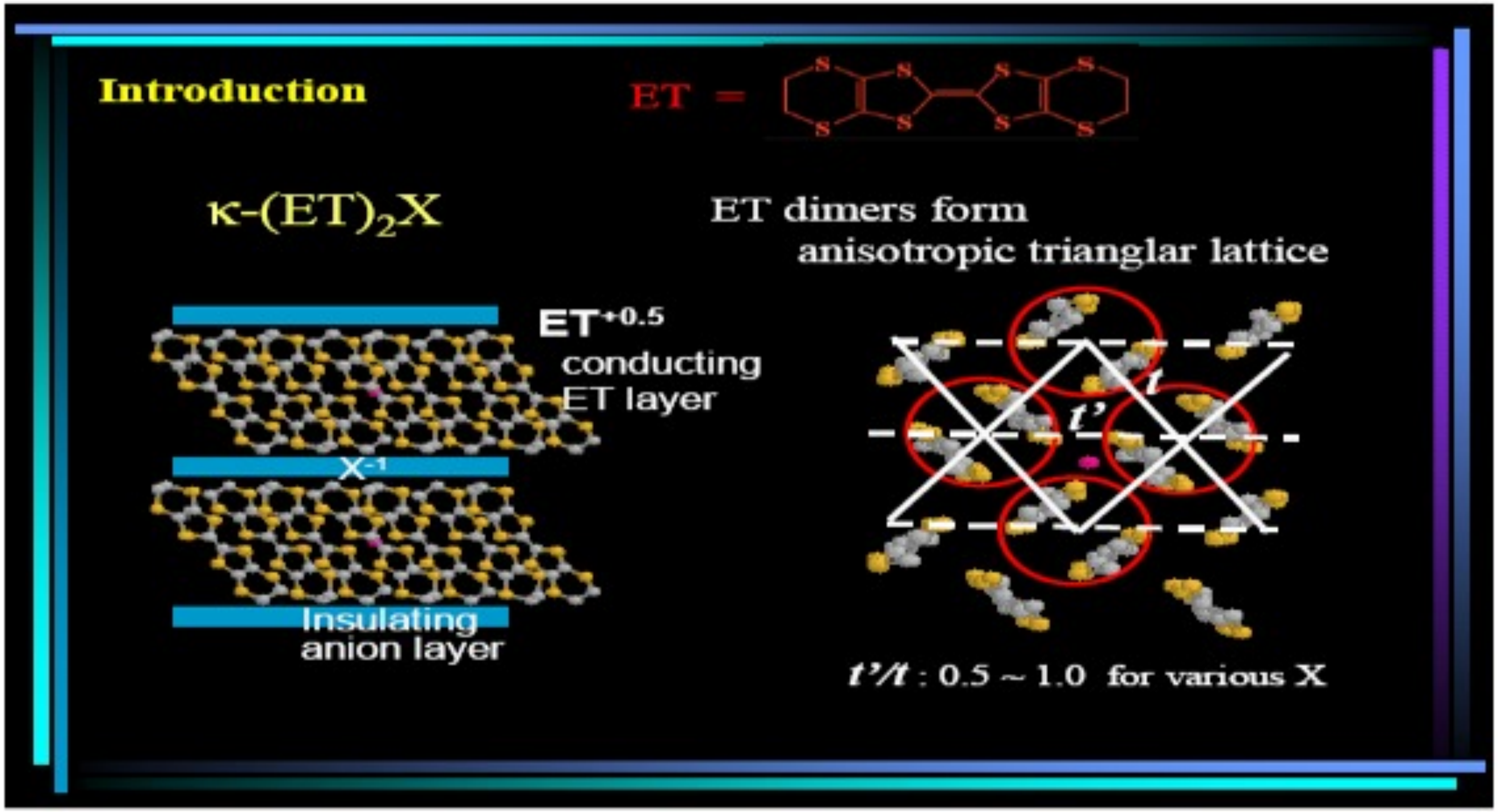
No magnetic ordering down to lowest measured T (<< natural exchange scales J)

BUT

Gapless excitations down to T << J.

Most extensively studied in organic spin liquids.

κ-(ET)$_2$X family of materials



Model by 1-band Hubbard model on anisotropic triangular lattice

No magnetic order

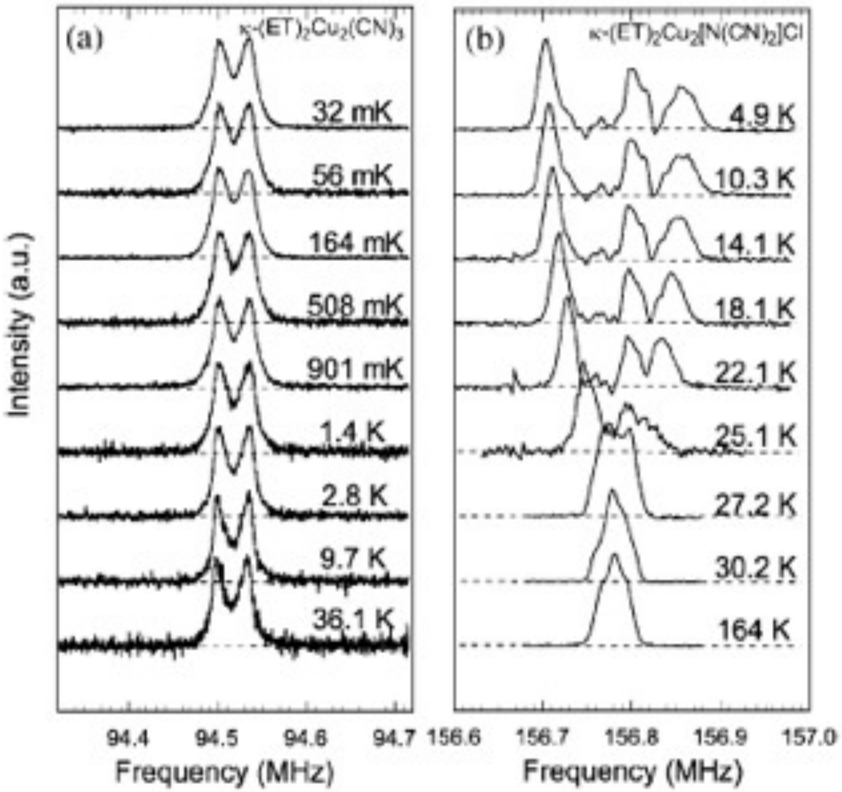
(Expt: Kanoda, 2002-present) κ-$(ET)_2Cu_2(CN_3)_2$

Weak Mott insulator close to Mott transition

$\approx$ Isotropic Δ lattice

No ordering to 32 mK

$\ll J \approx 250$ K

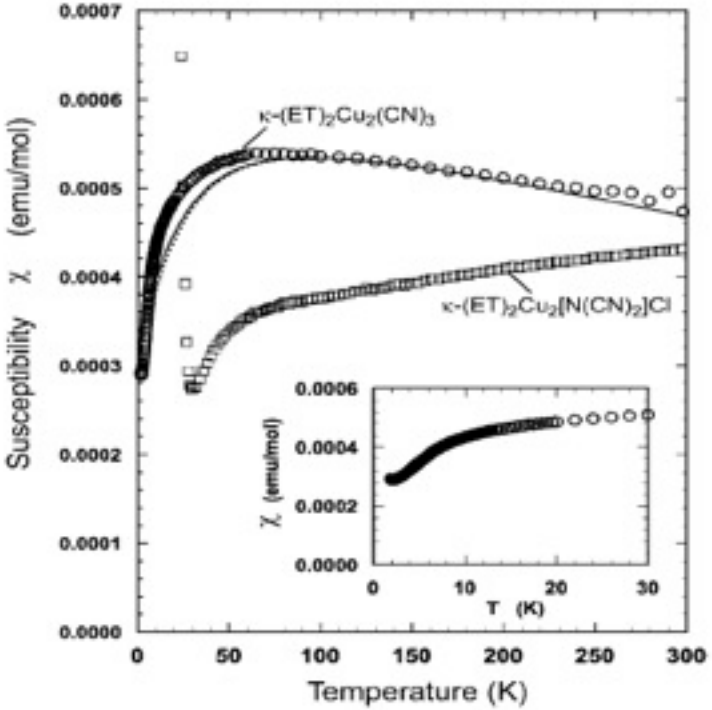


but $\chi \to$ const.

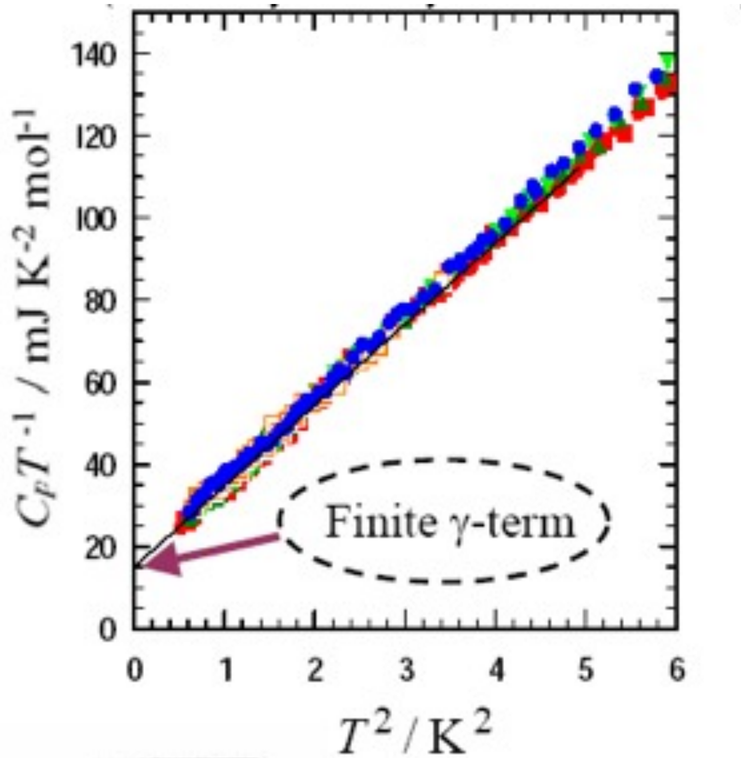
$\frac{1}{T_1 T} \to \frac{1}{\sqrt{T}}$ (above 10 K)

κ-E T:A gapless spin liquid
(at least down to 1 K << J ≈ 250 K)

Shimuzu, Kanoda et al, 2003



S. Yamashita et al, Nat Phys 08



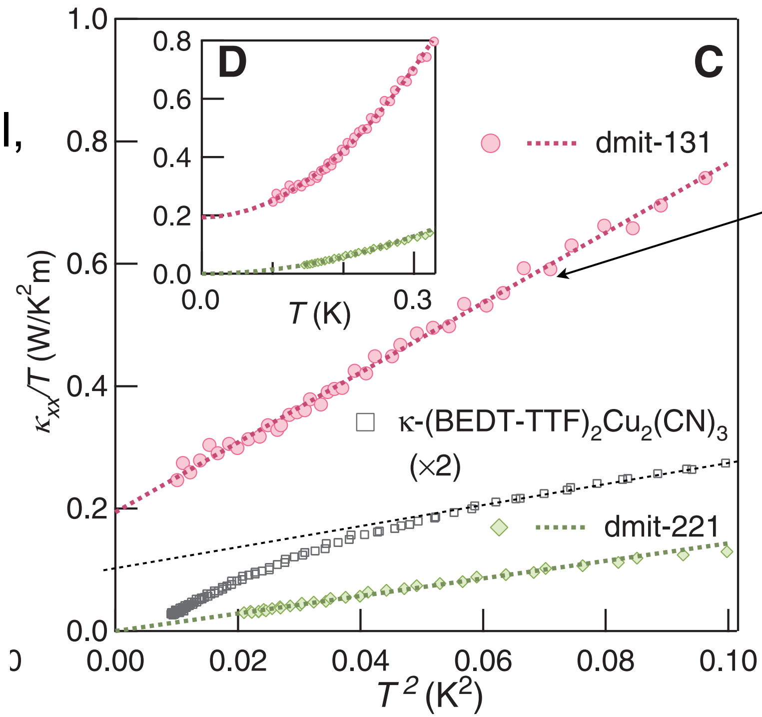
$\chi\ (T \to 0) \to$ const.

$C/T\ (T \to 0) \to$ const.

Wilson ratio $\frac{\chi T}{C}$ = const. $\sim O(1)$

Even more dramatic: Metallic thermal transport in a Mott insulator

M. Yamashita et al, Science 2010



dmit quantum spin liquid

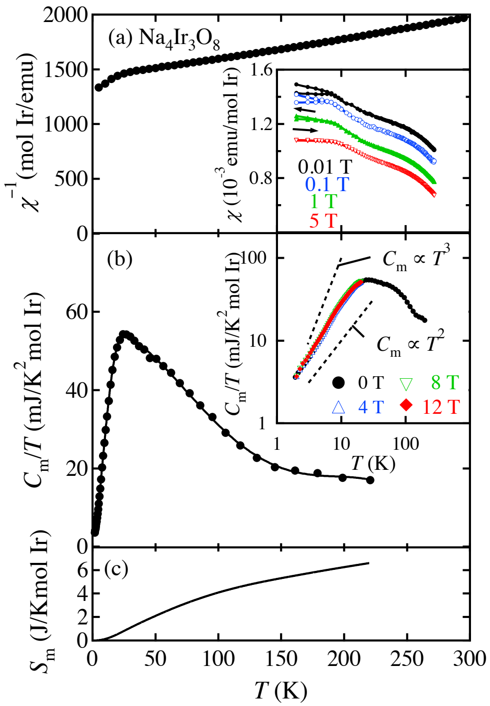
$$EtMe_3Sb[Pd(dmit)_2]_2$$

Gapless excitations are mobile in dmit spin liquid!
(More discussion of kappa-ET later).

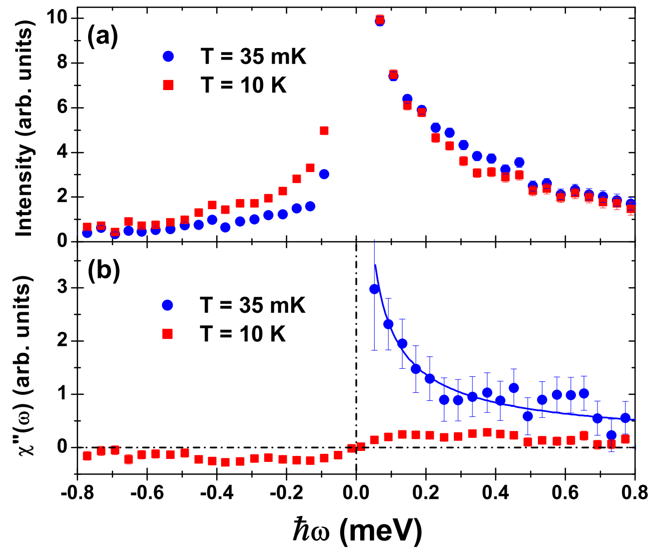
Other examples

$Na_4Ir_3O_8$ (3d hyperkagome)

$ZnCu_3(OH)_6Cl_2$ 2d Kagome



Okamoto et al, PRL 07



Helton et al, PRL, 07

Other nice examples: Volborthite , a distorted Kagome(Hiroi, Takigawa et al, 2007-present)

What about theory?

Quantum spin liquids may be either gapless or gapped.

Many distinct kinds of quantum spin liquid phases are theoretically possible (just like many different kinds of magnetic order).

Microscopic conditions which favor one or other of spin liquid states not understood in general.

Some rough insight

1. Weak Mott insulators:

Short distance physics is that of a metal but insulating at long distances - expect gapless spin liquids with fermionic `spinon' excitations

2. Strong Mott insulators with well developed short range magnetic order:

Short distance physics is that of an ordered magnet but no long range `phase coherence' - expect gapped spin liquids with bosonic spinon excitations.

However exceptions to these exist (apparently for instance in Kagome examples).

Questions for theory

1. Quantum spin liquids in weak Mott insulators?

2. Theoretical framework for gapless quantum spin liquids - some important ideas.

3. What experiments will most clearly reveal the physics?

4. Understanding existing experiments

5. How can cold atom experiments help?

Theoretical approaches to quantum spin liquids in a weak Mott insulator

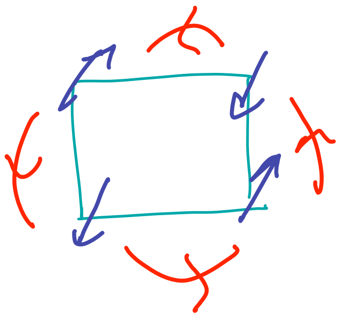
Approach from insulator

$t/U \nearrow$ $\Rightarrow$ Build in more virtual charge fluctuations in ground state wave function

$$H_{eff}[\{\vec{S}_i\}] = \sum_{ij} J_{ij}\, \vec{S}_i \cdot \vec{S}_j + K \sum_{\square} \left(P_{1234} + P^{-1}_{1234}\right) + \cdots$$

longer range exchange

Motrunich, 2005

P_{1234} =  = 4-particle ring exchange, etc

Ring exchange promotes spin liquids

Example: Ring exchange model on 2d triangular lattice

$$H = 2J_2 \sum_{\langle rr' \rangle} \vec{S}_r . \vec{S}_{r'} + J_4 \sum_{\square} (P_{1234} + \text{H.c.})$$

Many different theoretical methods:

Exact diagonalization (LiMing et al, 2000, H.-Y. Yang et al, 2010)

Variational wavefunctions (Motrunich 2005, ...)

DMRG, exact diagonalization on strips (Sheng et al, 2009).

Evidence for spin liquid behavior with increasing J4/J2.

Alternate: approach from the metal

Interacting Fermi fluid: Incorporate correlations with Jastrow factor

$$\psi_F(\mathbf{r}_1\sigma_1, ...\mathbf{r}_N\sigma_N) = \prod_{ij} f(\mathbf{r}_i - \mathbf{r}_j)\psi_{Slater}(\mathbf{r}_1\sigma_1, ...\mathbf{r}_N\sigma_N) \quad (1)$$

Special case: Gutzwiller approximation to lattice Hubbard model; choose

$$f_{ij} = g\delta_{ij} \quad (2)$$

with $g < 1$ to weigh down double occupancy of any site.

An interesting point of view

Can think of $\psi_F = (\text{Jastrow}) \times \psi_{\text{slater}}$

as $\psi_F = \underbrace{\psi_b(\vec{r}_1, \ldots, \vec{r}_N)}_{\text{Boson wave fn}} \; \psi_{\text{slater}}(\vec{r}_1\sigma_1, \ldots, \vec{r}_N\sigma_N)$

Clearly any choice of ψ_b will give a legitimate fermion wavefunction

Choosing ψ_b as wavefunction of superfluid leads to the Fermi liquid wavefunction ψ_F.

A interesting point of view (cont'd)

Spatial coordinates of bosons $\vec{r}_i$ are same as those of fermions irrespective of spin

$\Rightarrow$ (i) Bosons are "slaved" to the fermions.

(ii) Bosons should be viewed as spinless

Motivates a "slave boson" mean field theory of correlated metal

Slave boson mean field theory

Write electron operator as $c_{i\alpha} = b_i \, f_{i\alpha}$

b_i: spinless boson

$f_{i\alpha}$: spin-½ fermion ("spinon")

Replace microscopic H by equivalent approximate non-interacting H_{MF} for holons & spinons with self-consistently determined parameters

$$H_{MF} = \underbrace{H[b]}_{\text{Repulsive bosons}} + \underbrace{H[f]}_{\text{Non-interacting fermions}}$$

$$H[b] = -\sum_{ij} t^c_{ij} (b^\dagger_i b_j + h.c) + V_{int}[b^\dagger b]$$

$$H[f] = -\sum_{ij} t^s_{ij} (f^\dagger_{i\alpha} f_{j\alpha} + h.c)$$

f_α form a Fermi surface

Metallic phase: b condensed, $\langle b \rangle \neq 0$

$$\Rightarrow \quad c_{i\alpha} = \langle b \rangle f_{i\alpha}$$

Electron Green function $\langle c \bar{c} \rangle \simeq |\langle b \rangle|^2 \langle f \bar{f} \rangle$

$\Rightarrow$ Quasiparticle residue $Z = |\langle b \rangle|^2$

(= "condensate fraction" of boson)

Obtaining a Mott insulator from the metal

Start with wavefunction of correlated metal

$$\psi_f(\vec{r}_1\sigma_1, \cdots, \vec{r}_N\sigma_N) = \underbrace{\psi_b(\vec{r}_1, \cdots, \vec{r}_N)}_{\text{superfluid}}\,\psi_{Slater}(\{\vec{r}_\ell\sigma_\ell\})$$

How to get a Mott insulator?

Let $\psi_b \rightarrow$ wavefunction of localized solid of bosons

$\Rightarrow$ freeze out charge motion

$\psi_f = \psi_b^{solid}\,\psi_{Slater}$ is wavefunction for fermionic Mott insulator!

Comments

$\psi_F = \psi_b^{solid}\psi_{Slater}$ is a spin singlet wavefunction.
Expect spin correlations similar to a metal?

Other wavefunctions:

$\psi_F = \psi_b^{solid}\psi_{BCS}$ describes a different spin liquid state.
Spin correlations similar to a superconductor?

Extreme limit: Completely freeze out all charge fluctuations

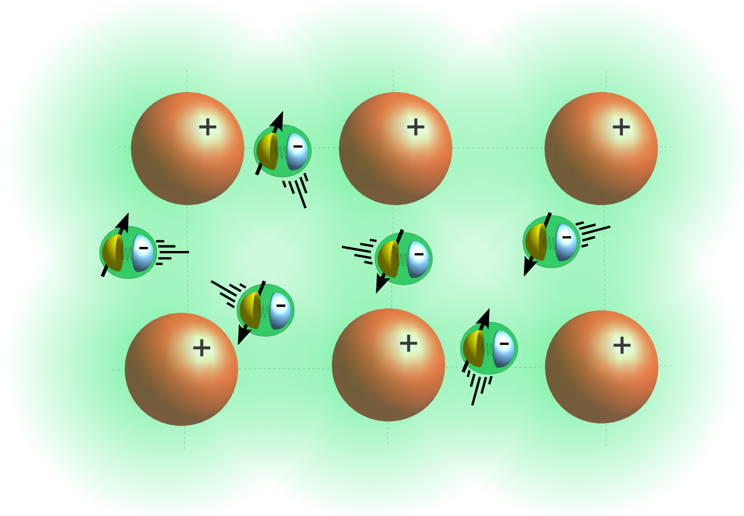
$$\psi_b^{solid} \rightarrow P_G \tag{1}$$

Gutzwiller projector $P_G = \prod_i (1 - n_{i\uparrow} n_{i\downarrow})$

Result: Pure spin wavefunction; can be tested variationally on ring exchange spin models derived in t/U expansion.

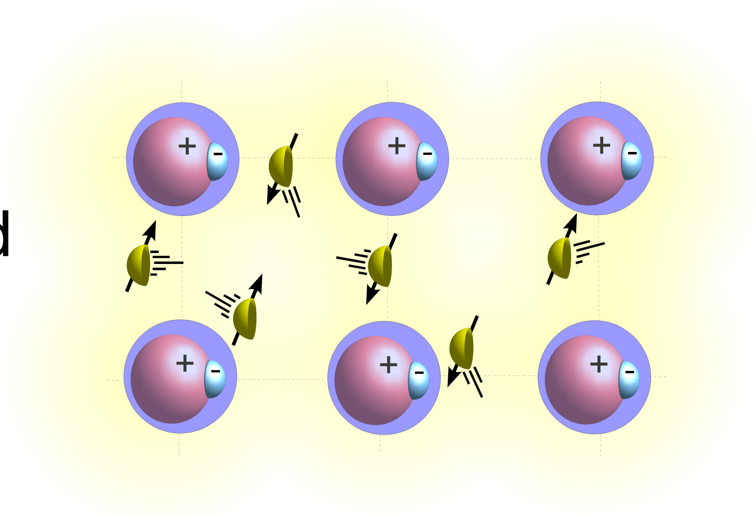
Picture of Mott transition

Metal



Electrons swimming in sea of +vely charged ions

Mott spin liquid near metal



Electron charge gets pinned to ionic lattice while spins continue to swim freely.

Formal theory

Slave boson mean field theory:

$$H_{mf} = H_b + H_f \tag{1}$$

$$H_b = -t_c \sum_{<ij>} \left(b_i^\dagger b_j\right) + U \sum_i \frac{n_i(n_i-1)}{2} \tag{2}$$

$$H_f = -\sum_{<ij>} t_{ij}^s \left(f_i^\dagger f_j + h.c\right) \tag{3}$$

Correlated metal: $t_c \gg U$, $< b > \neq 0$.

Mott insulator: $U \gg t_c$, bosons from a Mott insulator while fermions form a Fermi surface (i.e, a quantum spin liquid with spinon Fermi surface).

Readily generalize to other distinct quantum spin liquid states (eg BCS pairing of spinons).

Fluctuations: gauge theory

Slave particle representation $c_{i\alpha} = b_i f_{i\alpha}$

invariant under $b_i \to b_i e^{i\theta_i}$, $f_{i\alpha} \to f_{i\alpha} e^{i\theta_i}$

$\Rightarrow$ $U(1)$ "gauge" redundancy

$\therefore$ True low energy physics below charge gap

$$H = - \sum_{ij} t_{ij} \left(e^{i a_{ij}} f^{\dagger}_{i\alpha} f_{j\alpha} + h.c \right)$$

(+ constraint $\nabla \cdot E = f^{\dagger} f$)

Properties of this spin liquid (in d = 2)

Spinon Fermi surface + U(1) gauge field:

Large theoretical literature -

Holstein et al, 73, many papers in the 90s - RPA, large-N treatments

Some recent controversy (S.S. Lee, Metlitski, Sachdev): problems with the large-N expansion.

Resolution (Mross, McGreevy, Liu, TS, 2010): Combine large-N with another small parameter introduced by Nayak, Wilczek (94).

Controlled double expansion allows calculation of physics.

Properties of this spin liquid (cont'd) (in d = 2)

Specific heat $C_v \sim T^{\frac{2}{3}}$

Spin susceptibility $\chi \sim const$

Thermal conductivity $\kappa \sim T^{\frac{1}{3}}$.

Sharp $2K_f$ singularities in both spin density $f^\dagger \sigma f$ and *spinon density* $f^\dagger f$.

Meaning of gauge flux

Gauge flux density $b \sim \vec{S}_1 . \vec{S}_2 \times \vec{S}_3$ around a plaquette (scalar spin chirality).

Wen, Wilczek, Zee, 90; Lee, Nagaosa 91

Coupling to the internal gauge flux: Motrunich, 2007; also Chitra, Sen, 1990s

External B-field induces coupling to scalar spin chirality at $o(t^3/U^2)$

Therefore $b_{internal} = \alpha B$ is induced for the electrically neutral spinons.

α bigger in a weak Mott insulator.

Implication: B-field may have both orbital and Zeeman effects in weak Mott spin liquids!

Example: Thermal Hall effect (Katsura et al, 2010).

Properties of BCS paired spin liquids

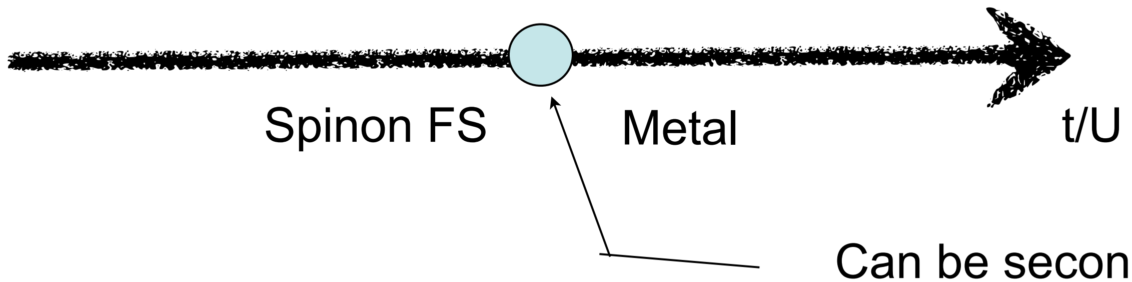
Spinon pair condensate expels U(1) gauge field.

Spin physics similar to that of corresponding paired superconductor
(eg: d-wave paired spinons => spin physics of d-wave BCS SC).

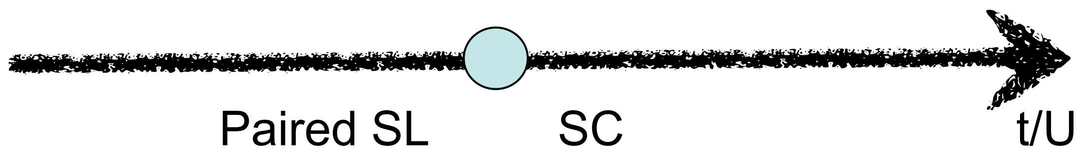
Vortices of spinon pair condensate: topological defects of the paired spin liquid

However for subtle reasons the vortices have a Z2 character (a vortex is its own antivortex): ``visons” (Ising-like vortex)

Quantum spin liquids and the Mott transition



Can be second order!
(TS,2008; Podolsky, Paremakanti, Kim, TS, 09)



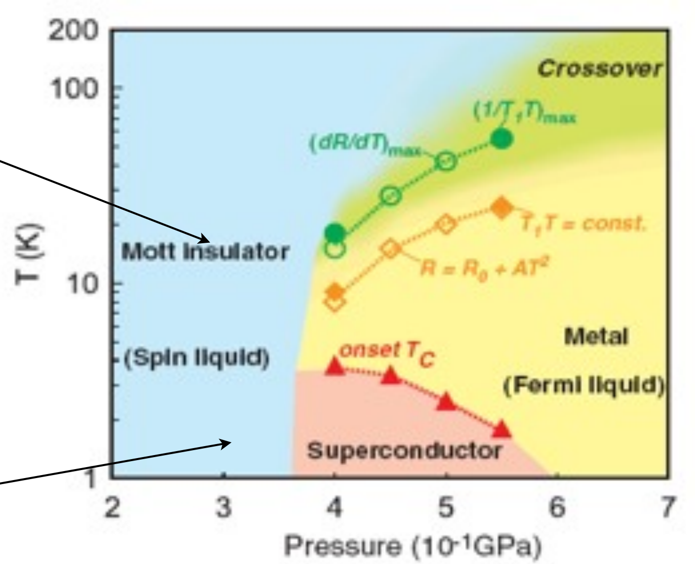
Application to experiment: Spinon FS as a universal intermediate temperature `mother' state

Spinon FS?

(Motrunich, 05)

Paired SL?

(Lee, Lee, TS, 06)



Low T instability in kappa-ET at ambient pressure at same temperature scale as SC instability under pressure

In dmit SL, no SC under pressure down to 1 K => weaker pairing tendency

Instability scale at ambient pressure also suppressed compared to kappa-ET

Crucial question

What experiments can reveal a `ghost' Fermi surface of spinons in the Mott insulator?

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What experiments can reveal a `ghost' Fermi surface of spinons in the Mott insulator?

A proposal (Mross, TS, 2010)

Charge Friedel oscillations:

- Kohn anomaly in phonon spectrum
- Standing wave patterns in STM for tunneling above the Mott gap

Physics of charge Friedel oscillations

Charge density correlations **at short distance** couple to spinon density correlations*.

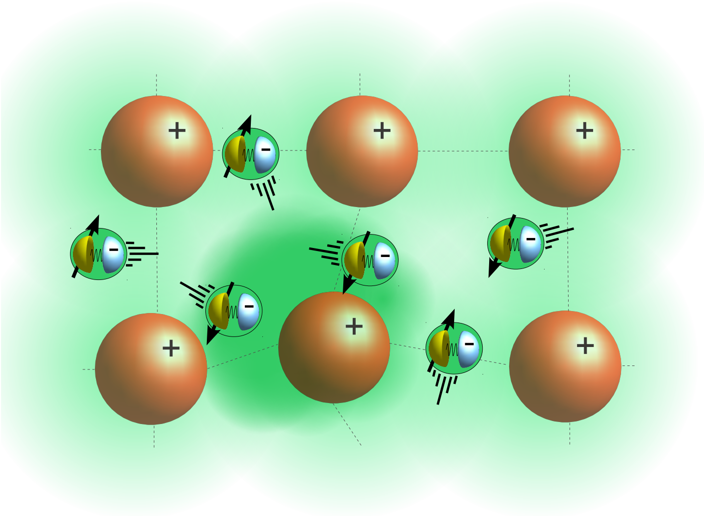
Spinon Fermi surface => spinon density correlations have sharp 2Kf singularities

=> Charge density correlations have 2Kf singularities in Mott insulator!

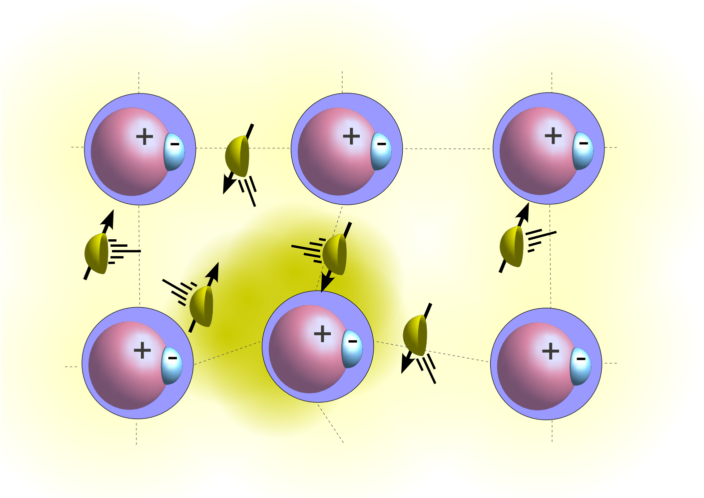
Mean field estimate: Magnitude unchanged across Mott transition but small compared to free Fermi gas.

*$\textbf{Spinon density}$ $f^\dagger f$ distinct from $\textbf{spin density}$ $f^\dagger \vec{\sigma} f$.

More useful: Kohn anomaly in phonon spectrum



Normal metal:
Ion motion screened by electron fluid;
Kohn anomaly due to change in screening at 2Kf wavevector



Spin liquid Mott insulator:
Ion bound to electron charge while electron spin stays mobile.

Ion-chargon motion carries gauge charge which is screened by spinon fluid => Kohn anomaly due to spinon FS.

Comments

1. 2Kf wavevectors known (approximately) for both organics, hyperkagome iridate.

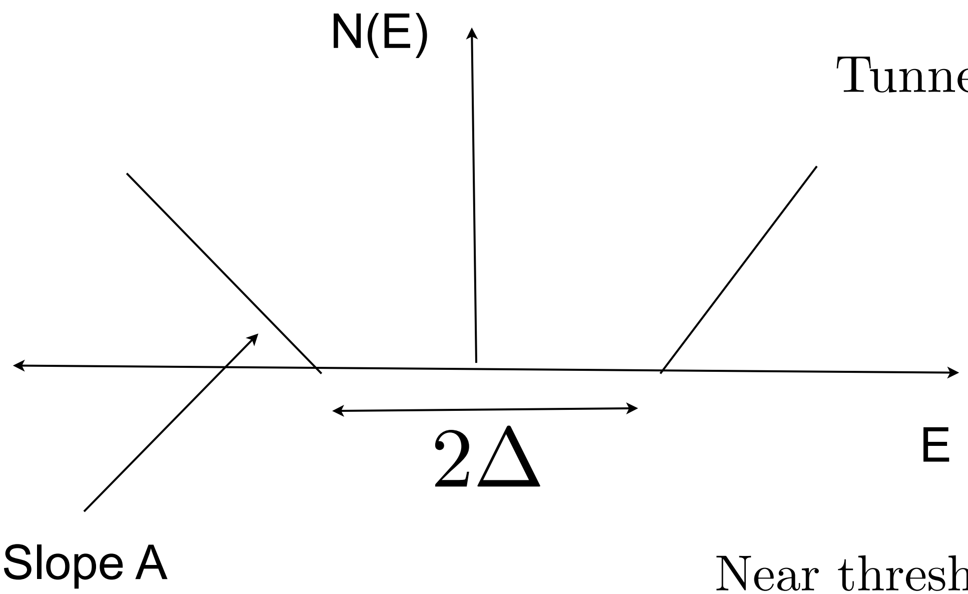
Obtain phonon spectrum thru inelastic X-ray?

2. Kohn anomaly survives even in strong Mott insulator if it has a spinon FS.

May be useful to look in Herbertsmithite, Volborthite, etc.

3. Phonon dynamics potentially useful probe of spinon physics in a gapless spin liquid Mott insulator.

STM to detect spinon Fermi surface in weak Mott insulators?



Tunneling conductance $\frac{dI}{dV} \propto N(E = eV)$

Tunneling requires injecting both the gapped charge and the gapless spinon => N(E) convolution of charge and spinon d.o.s

Near threshold $N(E) \approx A(|E| - \Delta)\theta(|E| - \Delta)$.

$\mathrm{A} \propto N_f(E = 0)$ (= spinon d.o.s at spinon Fermi surface)

=> near defects A = A(x) has spatial modulation at 2Kf wavevectors of spinon FS due to standing wave pattern of spinon d.o.s

=> study spatial modulation of A to determine 2Kf wavevectors.

Other ideas for detecting spinon Fermi surface

1. Quantum oscillations? (Motrunich 07)

Problems: unusual orbital response; low-T instability

2. Magnetic coupling of ferromagnets separated by spin liquid buffer (analagous to GMR) (Micklitz, Norman 09)

Problems: Cannot detect in resistivity, need atomic precision for spin liquid layer thickness

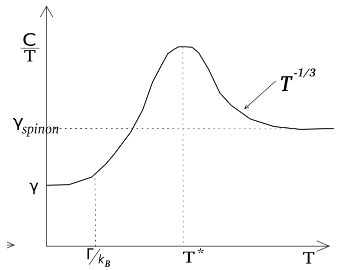
Low T instability in spin liquid organics: pairing instability of spinon FS?

Simplest option: $d_{x^2-y^2}$ pairing into a state with gapless nodal spinons.

Impurities - spin physics described by 'dirty d-wave' theory.

Grover, Trivedi, TS, Lee, 2010

Specific heat



Spin susceptibility $\chi \to const$, Wilson ratio ~ 1 confirmed by expt.

Predict metallic thermal conductivity $\kappa/T = const$ partially confirmed experiment on dmit.

Problem: field independence of low-T specific heat in expt.

Other pairing structure: Lee, Lee, TS, 2006; Galitski, Kim, 2007

Numerical evidence for d-wave spin liquid

Variational calculation for triangular lattice ring exchange model

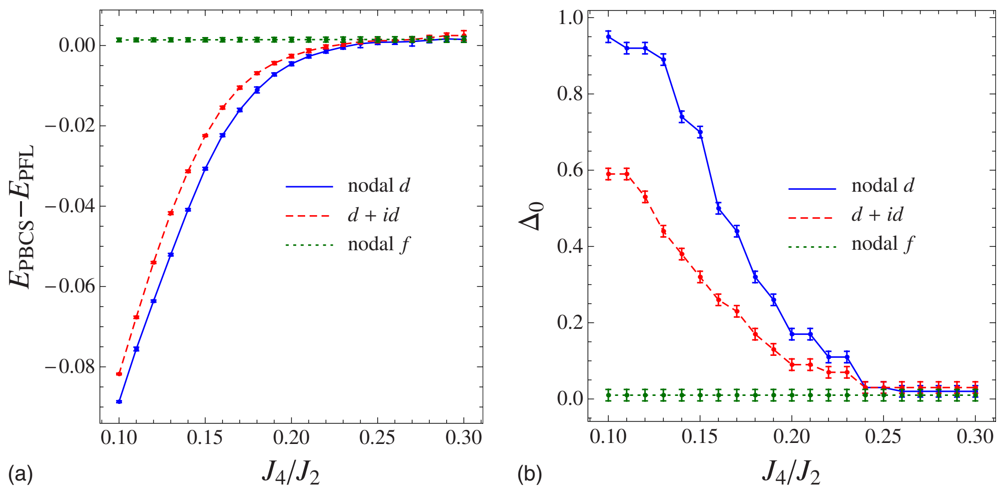
Grover, Trivedi, TS, Lee, 10

Compare SL wavefunctions

$|\psi_0\rangle = P_G |FL\rangle$

$|\psi\rangle = P_G |BCS\rangle$ with various pairing symmetries

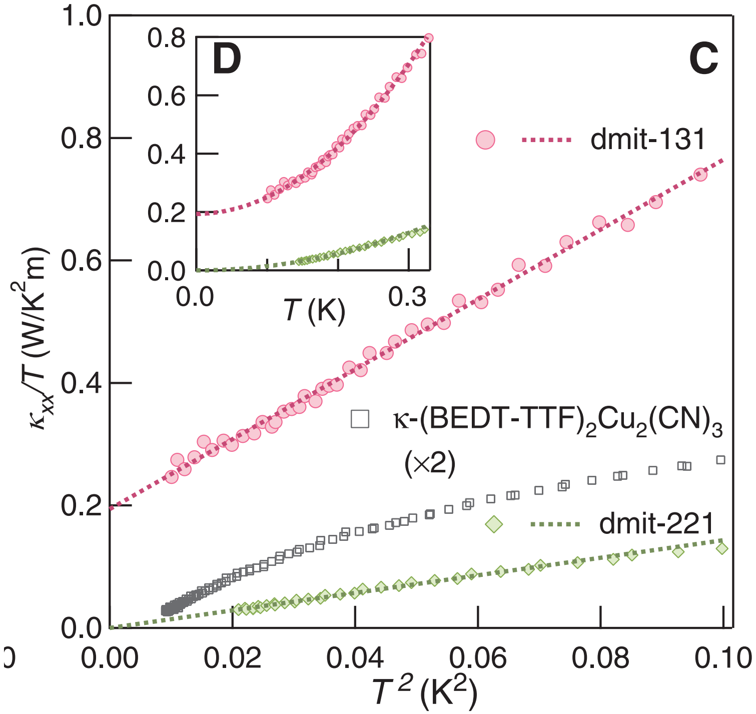
$$H = 2J_2 \sum_{\langle rr' \rangle} \vec{S}_r \,.\, \vec{S}_{r'} + J_4 \sum_{\square} (P_{1234} + \text{H.c.})$$



Projected FL wins for large J4 but nodal d-wave wins for intermediate J4.

Nodal d-wave: break lattice rotation but not translation => nematic spin liquid

Thermal conductivity in organic spin liquids

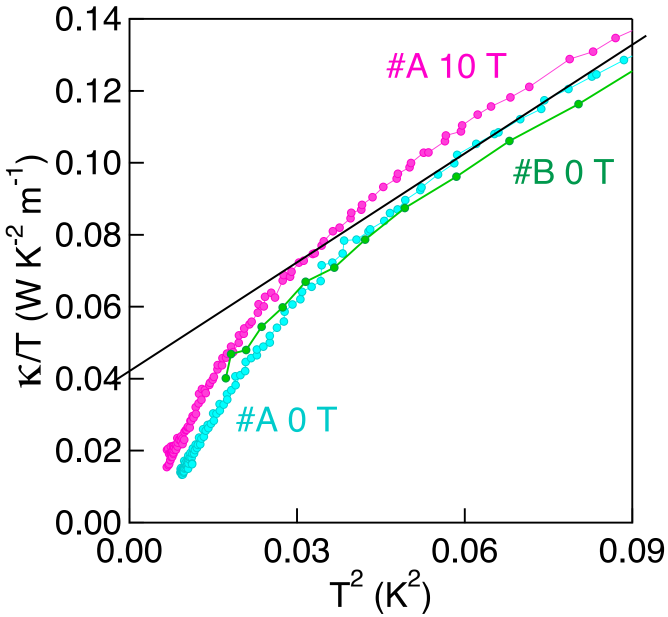


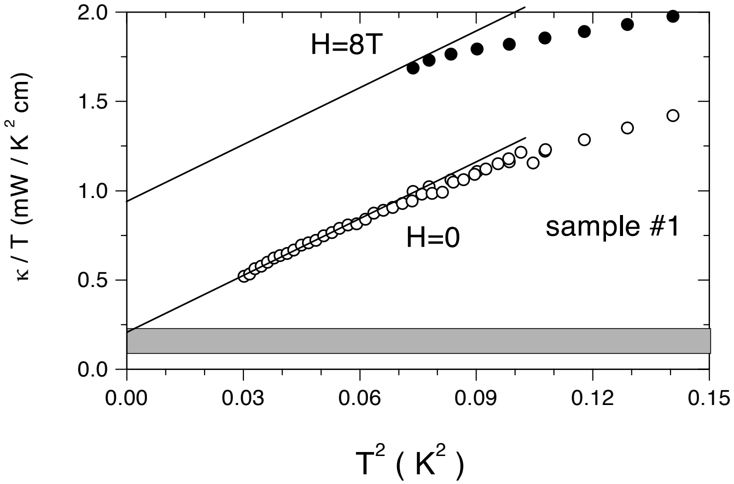
Huge residual heat conductivity in dmit

Within "dirty d-wave" theory, $\frac{v_F}{v_\Delta} = 550$
(compare with $\frac{v_F}{v_\Delta} = 14$ for optimal YBCO, $= 280$ for overdoped Tl-2201

v_F: velocity normal to FS
v_Δ: velocity parallel to FS

Thermal transport in kappa-ET SL - compare with kappa-ET dSc





Spin liquid $\kappa-(ET)_2Cu_2(CN)_3$

M. Yamashita et al, Nat. Phys. 08

d-wave SC $\kappa-(ET)_2Cu(NCS)_2$

Behnia et al, PRL, 1998

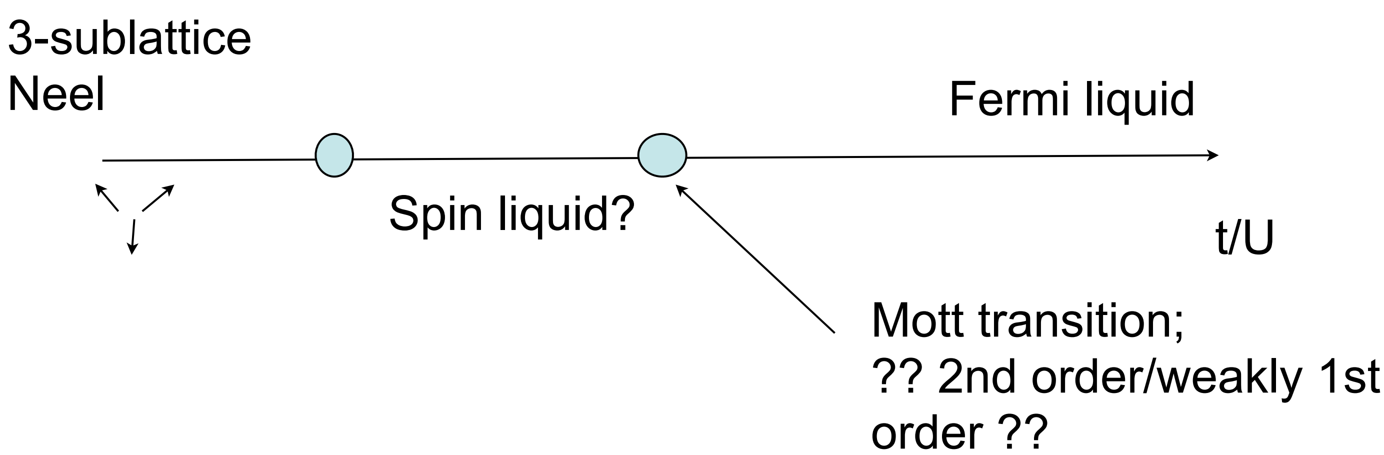
Very similar data above 0.2 K

How can cold atom experiments help?

Short term:

Study Mott transition in Hubbard model of cold spinful fermions on triangular lattice at **not too low temperature.**

Phenomenology more interesting than in square or cubic lattice.

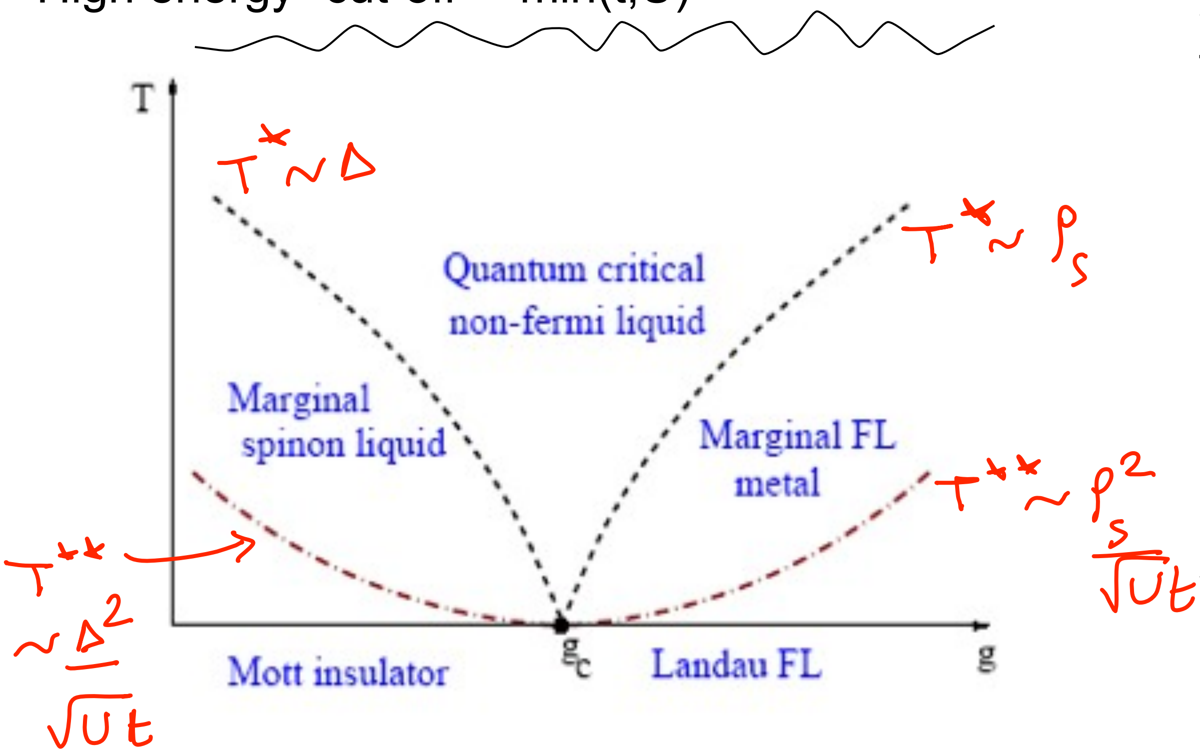


Saturday, September 25, 2010

Expected finite-T phase diagram

TS, 2008

High energy `cut-off' ~ min(t,U)



J not small close to Mott transition.

IF Mott transition is second order/weakly first order........

Do not may be need to cool to very low-T to see interesting physics (i.e signs of spin liquid)

Expected finite-T phase diagram

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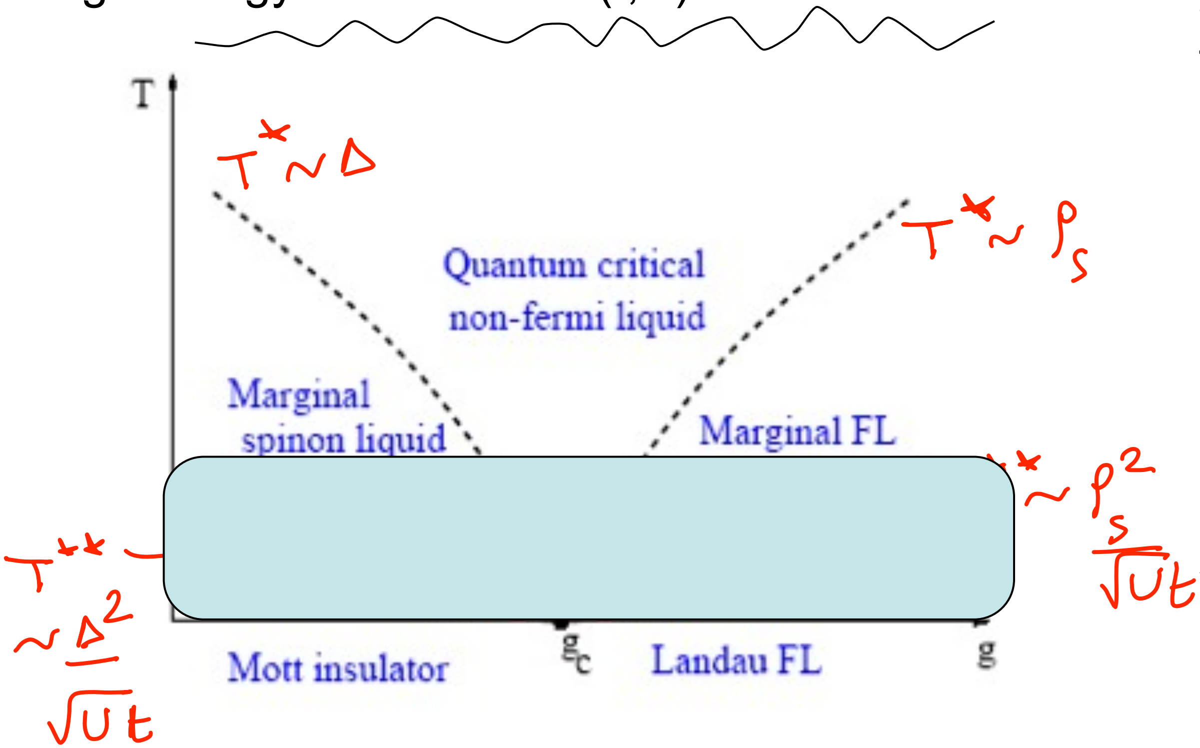

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Summary

Quantum spin liquid states discovered in experiments in last few years.

Growing number of experimental candidates - many dramatic phenomena.

All experimental candidates are gapless (at least to very low T).

Theoretical framework: Spinon FS at intermediate temperature, instability (pairing?) at very low T.

Needed: experimental detection of spinon FS, gauge field effects.

Very low-T state seen in experiments remains to be clarified.

Cold atom experiments: Studying vicinity of Mott transition in fermionic Hubbard model at not too low T is feasible and will be very useful!!