Quantum spin liquids and the Mott transition

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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.

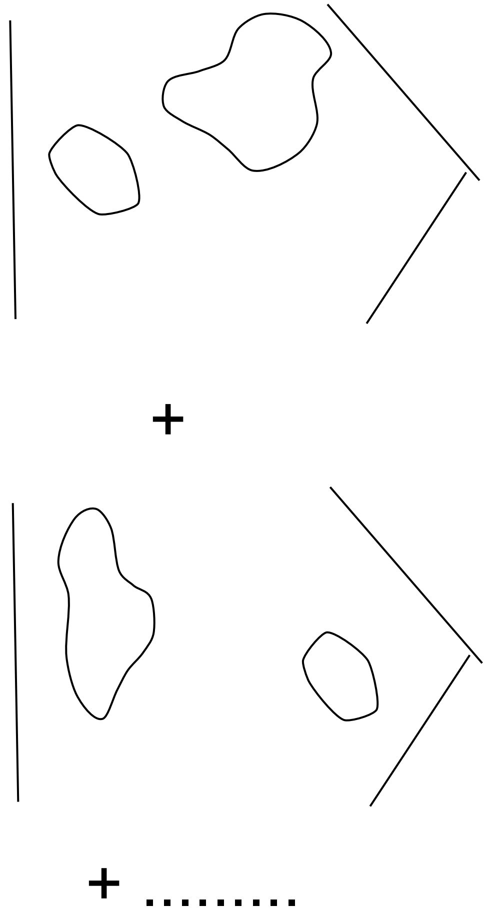
1930s- present: elaboration of broken symmetry and other
states with short range entanglement

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

Some candidate materials

$\kappa - (ET)_2 Cu_2 (CN)_3$

$EtMe_3Sb[Pd(dmit)_2]_2$

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

$Na_4 Ir_3 O_8$

Three dimensional `hyperkagome' lattice

$ZnCu_3(OH)_6Cl_2$

Volborthtite, ...

2d Kagome lattice (`strong' Mott insulator)

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Three dimensional `hyperkagome' lattice

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

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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 with $J \approx 250$ K.

Plan for talk

1. Brief discussion of a theoretical framework for spin liquids in weak Mott insulators.

2. Some facts and simple theory at low-T in the organics

3. Proposals for future experiments.

Theoretical approaches to quantum spin liquids in a weak Mott insulator

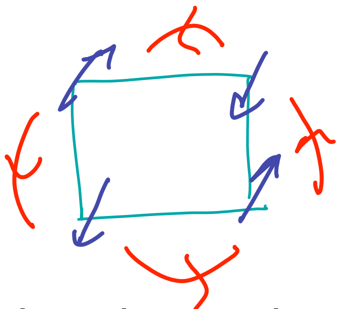
Approach from insulator

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

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

longer range exchange

Motrunich, 2005

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

Various numerics: ring exchange promotes spin liquids (LiMing et al 00, Motrunich 05, H.-Y. Yang et al, 2010)

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

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

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.

Obtaining a Mott insulator from the metal

Start with wavefunction of correlated metal

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

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

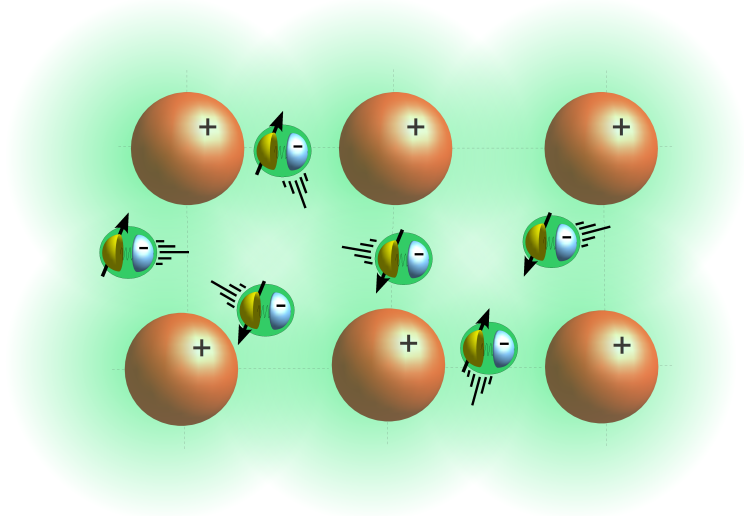
$$\psi_b^{solid} \to P_G \tag{1}$$

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.

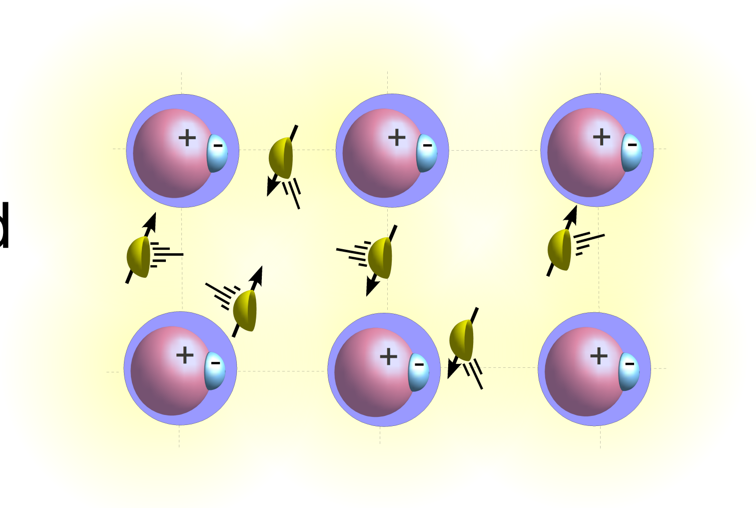
Picture of Mott transition

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 particle representation:

$$c_\alpha = b f_\alpha$$

b: charge-e spin-0 boson (chargon/holon)

f: charge-0 spin-1/2 fermion (spinon)

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 (cont'd) (in d = 2)

RPA theory: many papers in the 90s;
Recent controlled calculation beyond RPA: Mross, McGreevy, Liu, and TS, 2010.

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*.

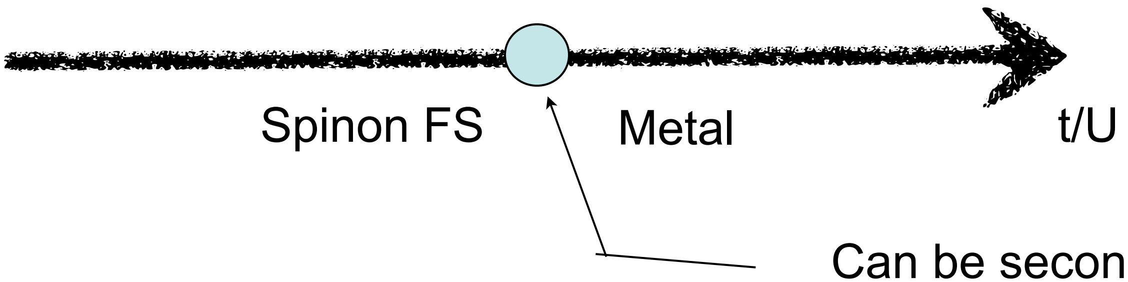
Spin physics similar to that of corresponding paired superconductor (eg: d-wave paired spinons => spin physics of d-wave BCS SC).

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

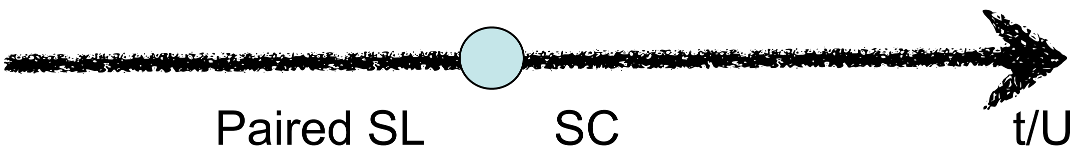
However for subtle reasons the vortices have a Z2 character (a vortex is its own antivortex): ``visons" (Ising-like vortex)

* => no simple orbital effect of external B-field, no simple prediction for thermal Hall effect.

Quantum spin liquids and the Mott transition



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



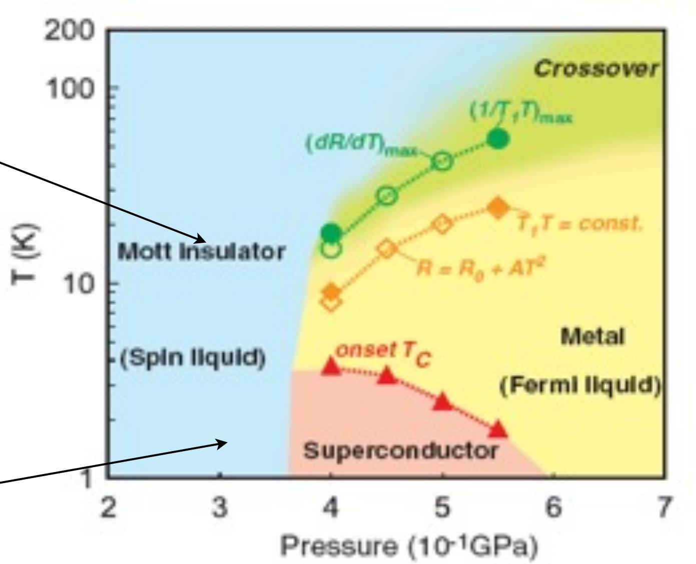
Application to experiment: Spinon FS as a universal intermediate temperature `mother' state

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

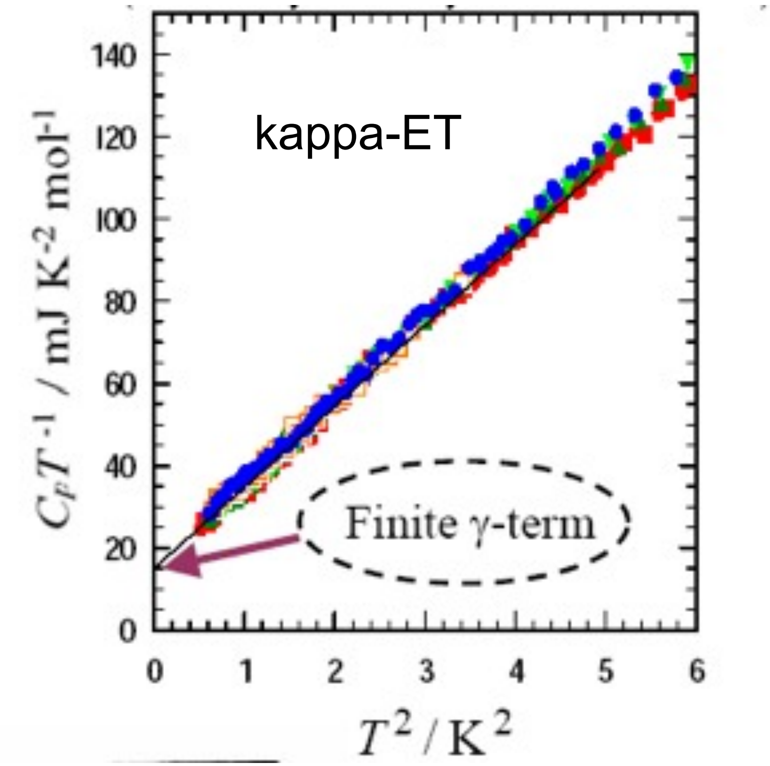
Instability scale at ambient pressure also suppressed compared to kappa-ET

Low-T physics of organic spin liquids: some facts and some simple theory.

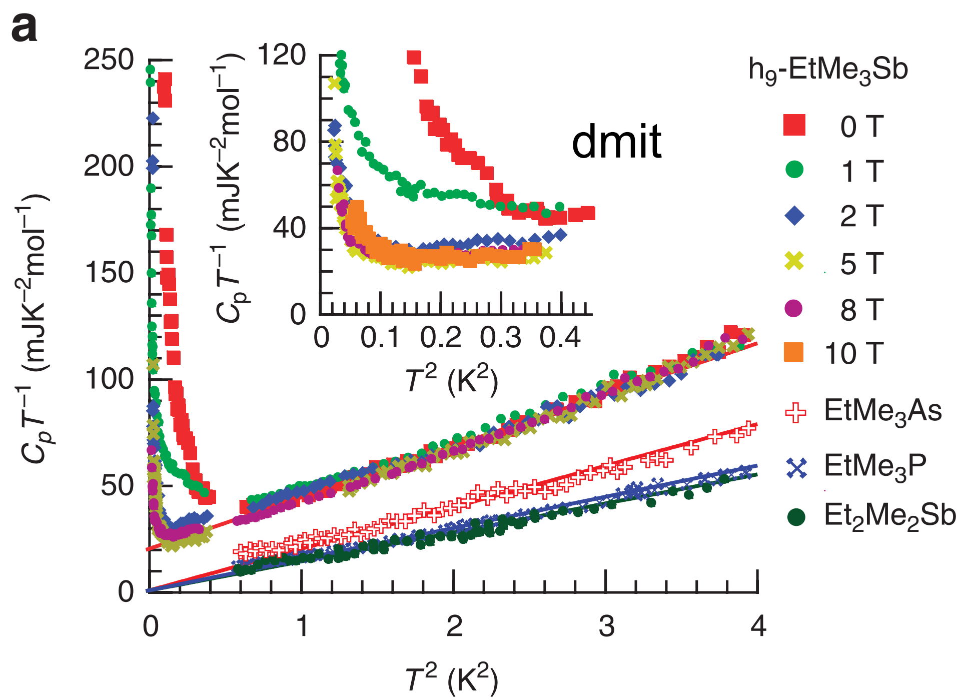
Low-T (ambient pressure): gapless excitations

Linear-T specific heat in a Mott insulator!

$$c_p = \gamma T \text{ at low-T}$$



S. Yamashita et al, Nat Phys, 08

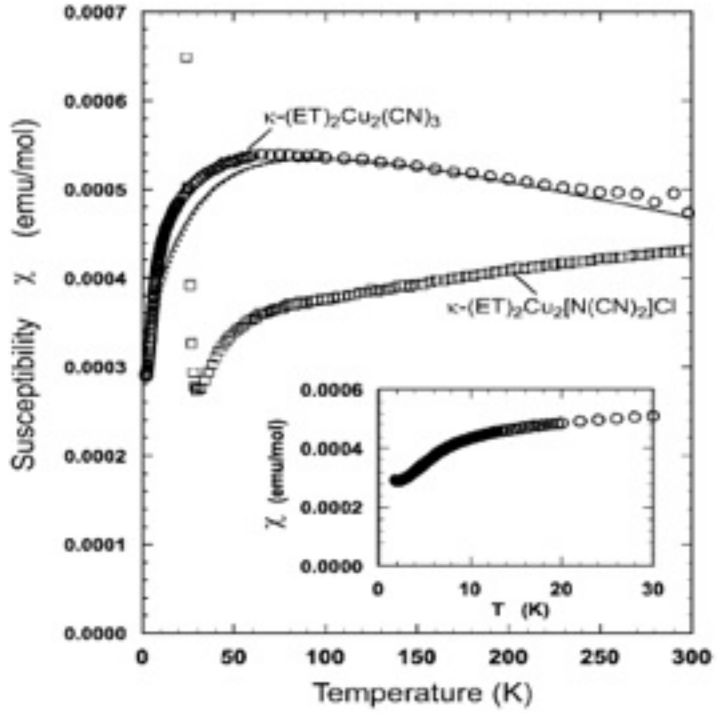


S. Yamashita et al, Nat. Comm., 2011

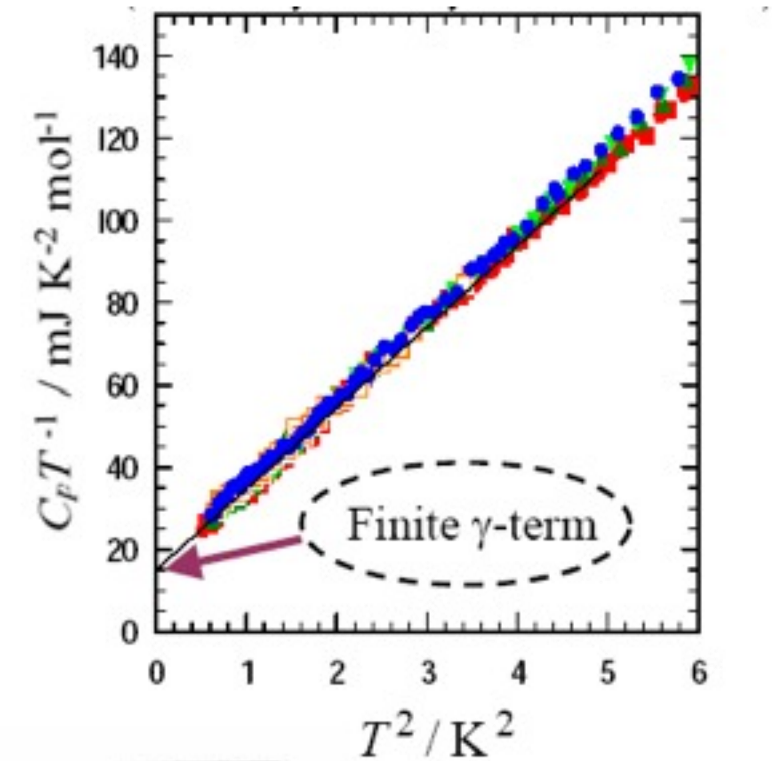
Data extends to < 1 K << J = 220 - 250 K.

Do gapless excitations carry spin? Example κ-E T (similar in dmit)

Shimuzu, Kanoda et al, 2003



S. Yamashita et al, Nat Phys 08



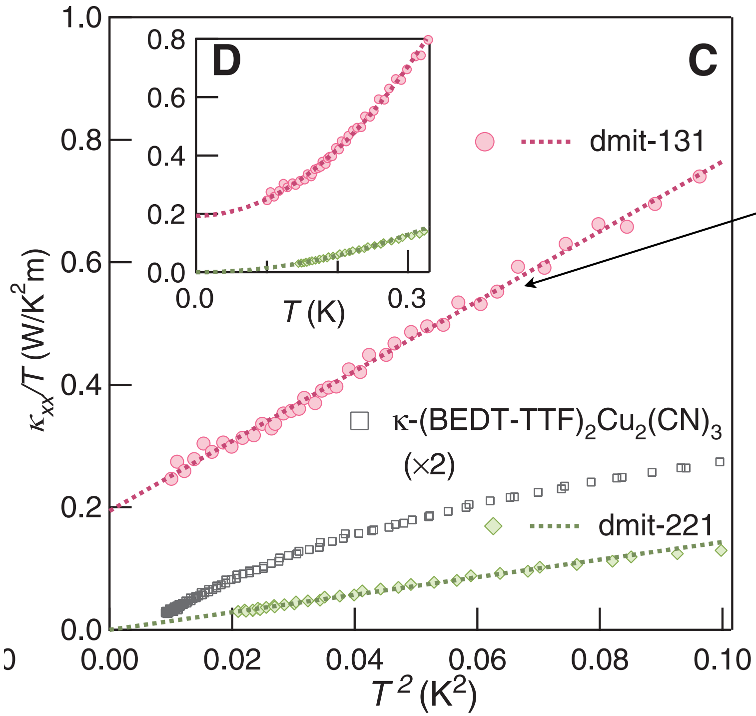
$\chi\,(T \to 0) \to$ const.

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

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

Are gapless excitations mobile?

M. Yamashita et al, Science 2010



dmit quantum spin liquid

Dramatic result - metallic thermal transport in a Mott insulator.

$\kappa = \frac{1}{3} C v l$

Estimate velocity $v \approx Ja$ to get mean free path $l \approx 50a \approx 500A$.

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

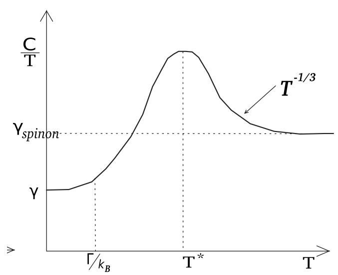
Low T instability in spin liquid organics: pairing instability of spinon FS?

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.

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

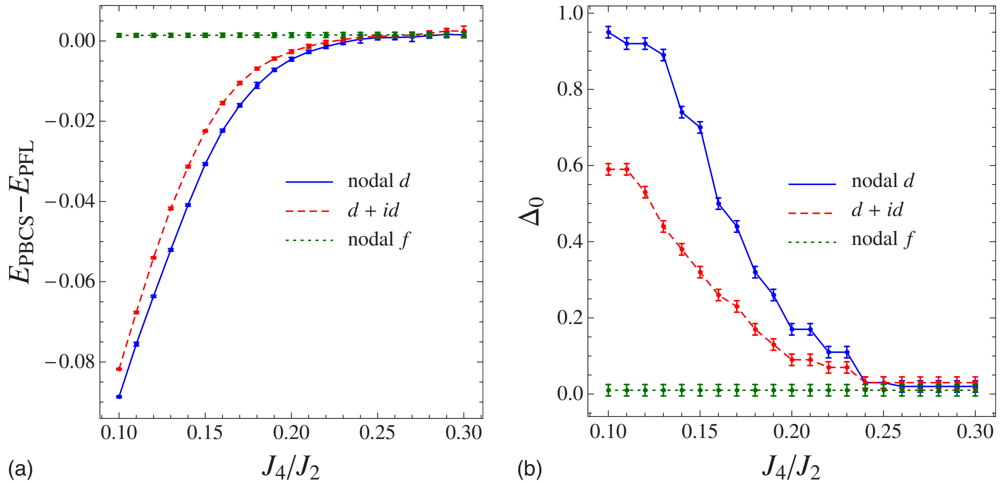
Grover, Trivedi, TS, Lee, 10

Compare SL wavefunctions

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

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

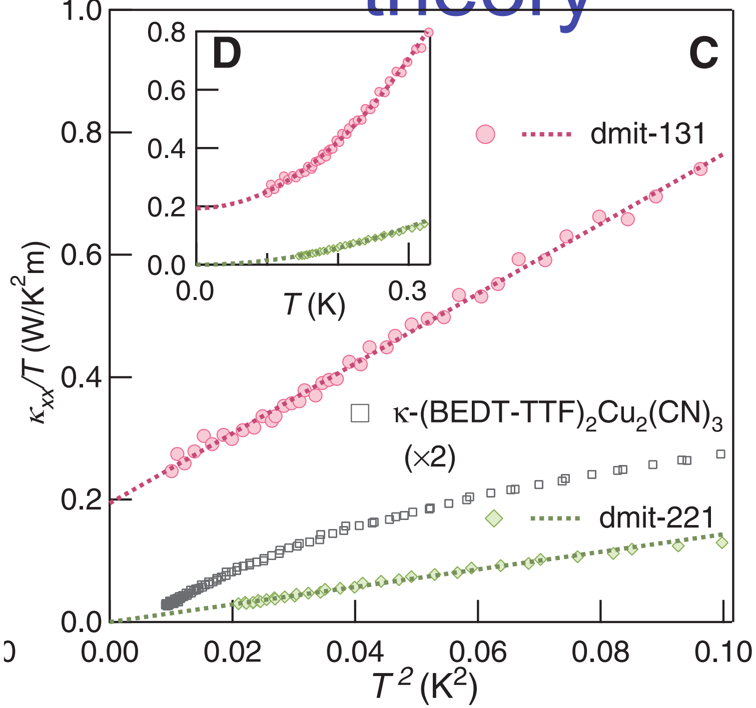
$$H = 2J_2 \sum_{\langle rr'\rangle} \vec{S}_r \,.\, \vec{S}_{r'} + J_4 \sum_{\square} (P_{1234} + \text{H.c.})$$



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 dmit: dirty d-wave theory

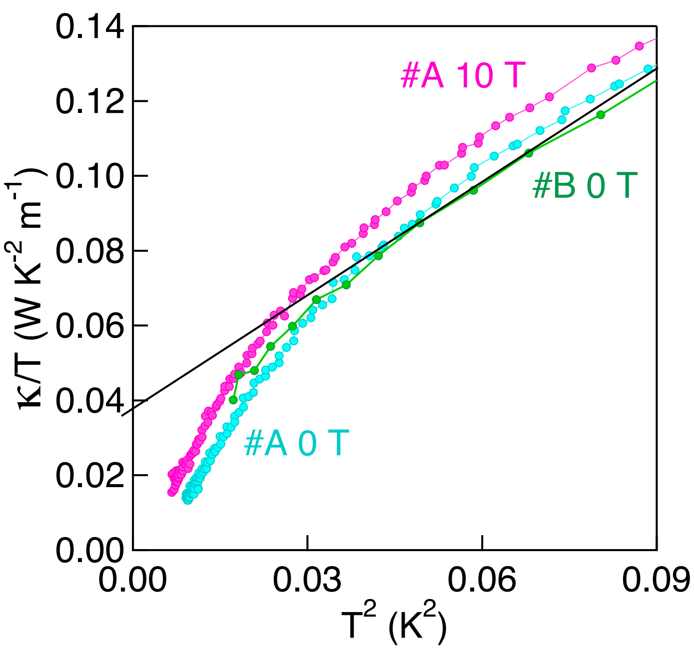


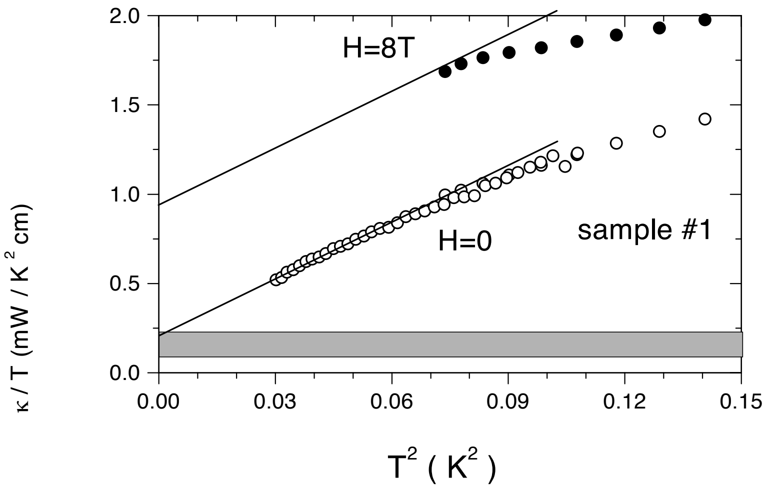
Huge residual heat conductivity in dmit

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

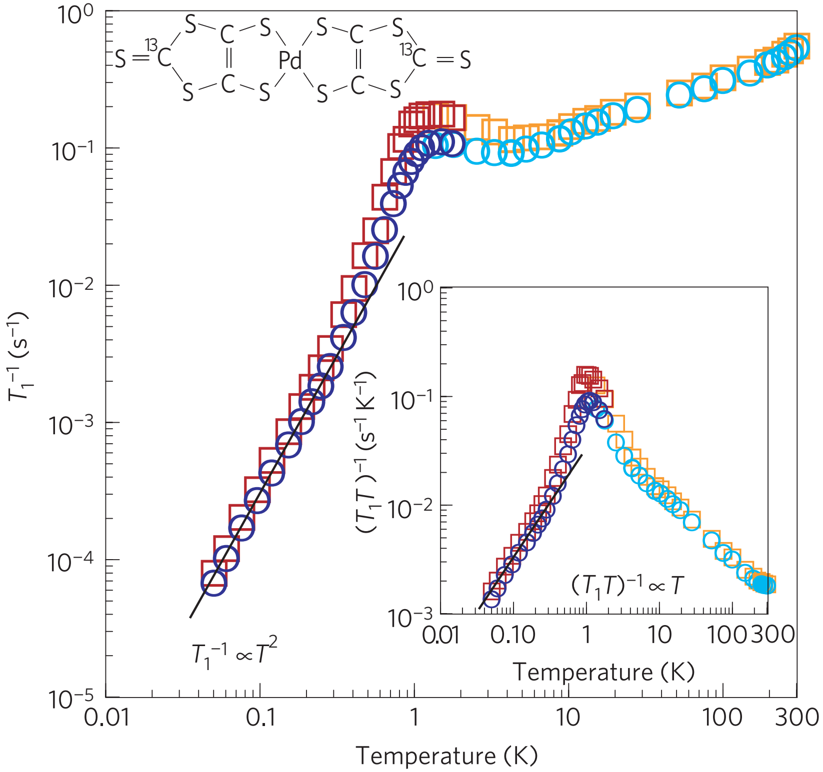
d-wave SC $\kappa-(ET)_2Cu(NCS)_2$

Behnia et al, PRL, 1998

Very similar data above 0.2 K

Origin of very low-T downturn - an eventual small gap? loss of thermal contact with spins?

Puzzles from NMR in dmit



T-dependence not expected for fermionic spin carriers with constant density of states.

In same T-H range get metallic thermal transport.

Puzzle: why is whatever is carrying the heat apparently not able to relax the nuclear spin?

Itou et al, Nat Phys 2010

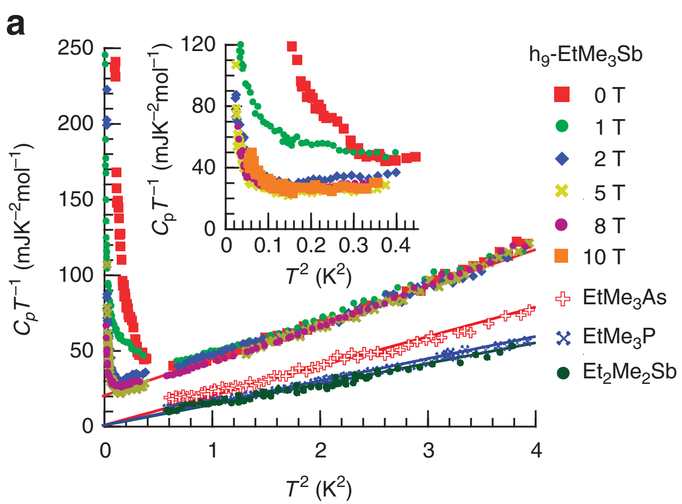
Suggestions in literature: May be gapless excitations are spinless `Majorana' fermions, other more exotic.

A resolution

TS, unpublished

Estimate expected NMR relaxation rate based on measured density of states of mobile fermions and available estimates of hyperfine coupling.

$$\frac{1}{T_1} \approx \frac{\pi \hbar k_B T}{2} (A\mu_B)^2 \left(\rho(E_f)\right)^2$$



$\gamma = 20 mJ/molK^2$ gives density of states

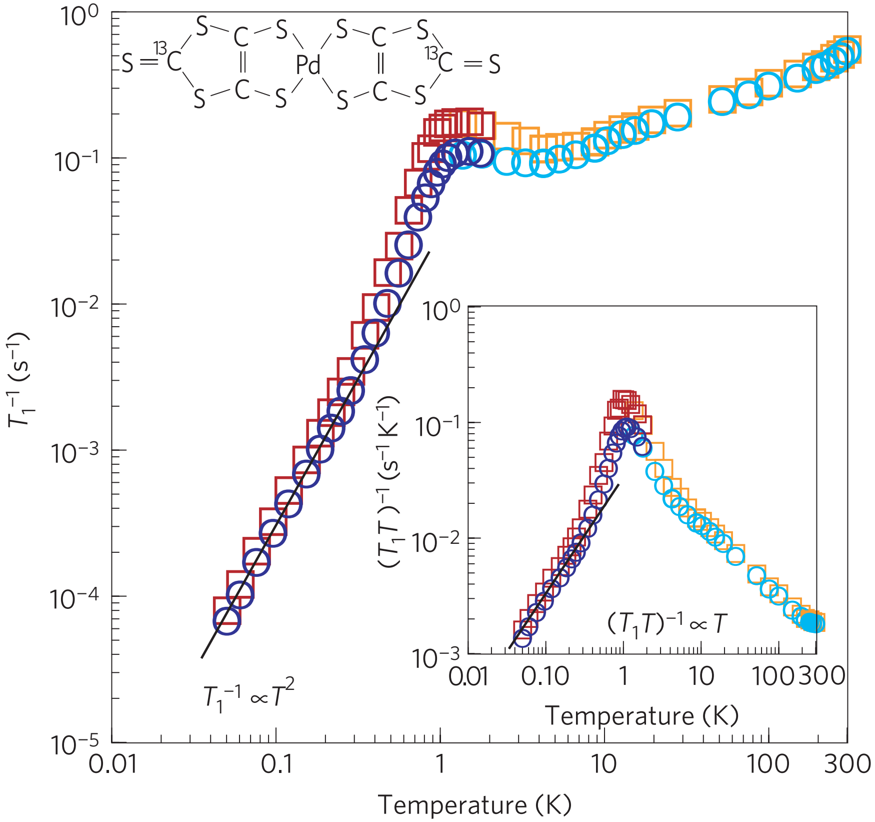
Hyperfine coupling $A \approx 900 kHz/\mu_B$ (high-T NMR, etc)

Itou et al, 08

$$\frac{1}{T_1 T} \approx 10^{-5} s^{-1} K^{-1}$$

Measured $\frac{1}{T_1 T}$ is much **bigger** than the estimated one!

TS, unpublished



~~Puzzle: why is whatever is carrying the heat apparently not able to relax the nuclear spin?~~

Real puzzle: why is nuclear spin relaxing so quickly?

Some possibilities

1. Spinon fermi surface has enhanced 2Kf spin fluctuations compared to Fermi liquid => enhanced NMR relaxation.

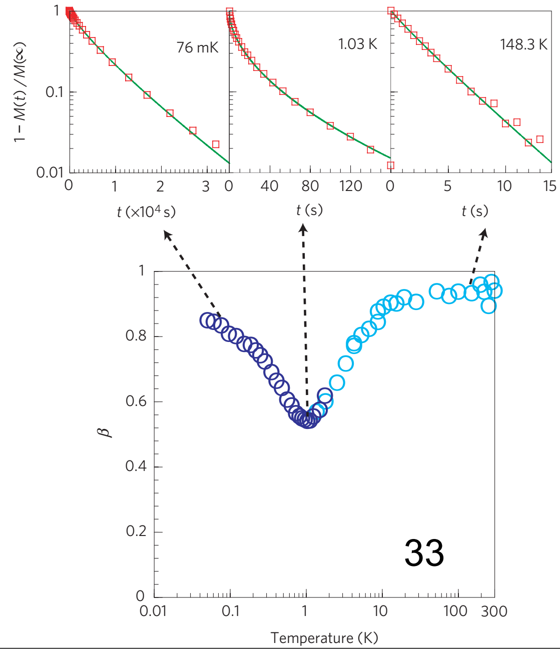
Pairing at low-T => gradual suppression of this enhanced 1/T1.

2. Competition between spinon pairing and antiferromagnetism.

Enhanced AF fluctuations above pairing transition which are suppressed once pairing occurs.

Caution: Around 1 K, NMR relaxation is not single exponential => distribution of relaxation times;

Presence of inhomogenous component possibly related to magnetism?



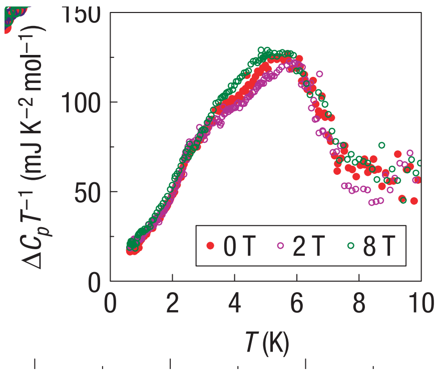
Comments on d-wave paired nodal nematic spin liquid

1. Spin excitations: (a) Gapless nodal spinons
(b) Gapped spin-1 resonance (analogous to famous neutron resonance in cuprates, etc)

- possibility of field-induced antiferromagnetism coexisting with nodal spin liquid.

Explanation of field-induced staggered moment seen in NMR (Shimuzu et al, 2004, Itou et al 2008) and muSR (Pratt et al 2011) ?

2. Difficulty: field independence of low-T specific heat



Theory not perfect but less imperfect than other ideas one can explore!

Fundamental theoretical concept: Spinon Fermi surface at intermediate-T.

Basic framework for thinking about low-T physics (instability of spinon fermi surface).

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*.

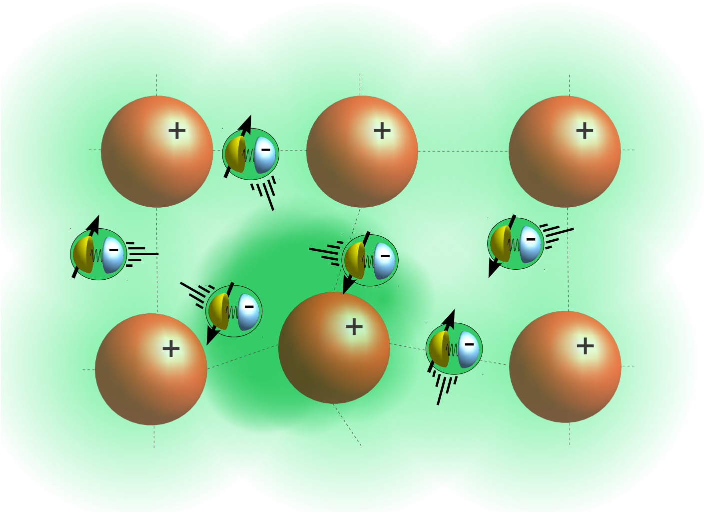
Spinon Fermi surface => spinon density correlations have sharp 2Kf singularities

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

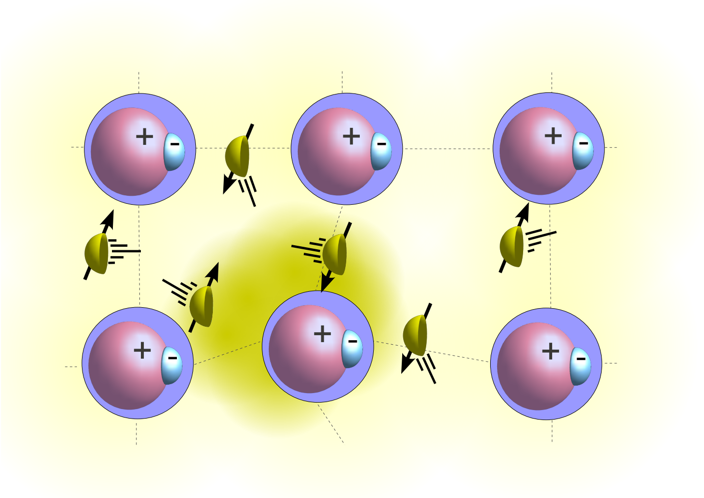
Mean field estimate: Magnitude unchanged across Mott transition but small compared to free Fermi gas.

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

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.

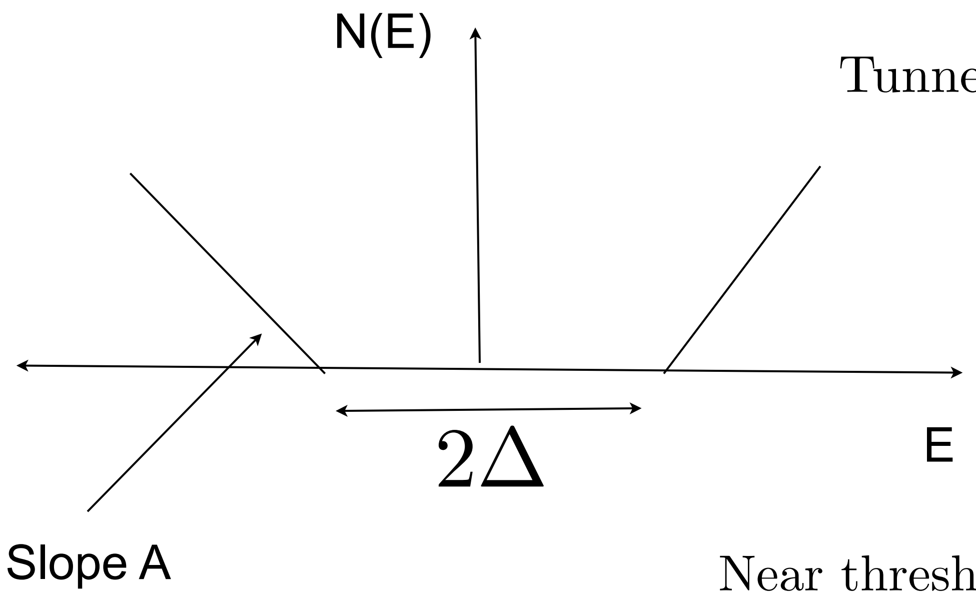
Obtain phonon spectrum thru inelastic X-ray?

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.

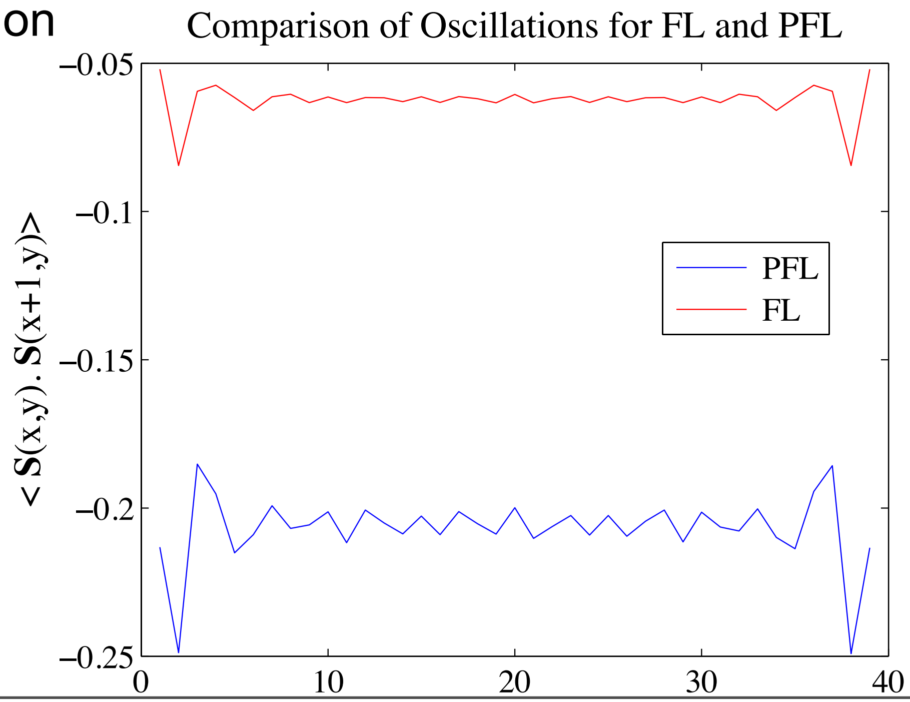
Interpretation of Friedel oscillations

Impurity in this spin liquid leads to oscillations of bond energies at 2Kf wavevectors.

``Impurity-pinning" of incommensurate valence bond order.

Demonstrate in a projected wavefunction calculation (T. Grover, unpublished).

Compare bond energy near step edge in Fermi Liquid (FL) and spin liquid described as Gutzwiller Projected Fermi Liquid (PFL)



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

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.