Quantum spin liquid:—A novel quantum state of matter—

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OUTLINE

1. Introduction

2. A possible quantum spin liquid state in the 2D organic Mott insulators with triangular lattice

3. Elementary excitations and phase diagram of the quantum spin liquid

4. A novel quantum phase in the Mott Insulator
   Can insulators have Fermi surface?

5. Summary

Quantum Liquid

Classical Liquids

- Frozen at the absolute zero temperature
- Ordering in conventional way, e.g., crystallization

Quantum Liquids

- No freezing by quantum fluctuation

Zero point oscillation > Interaction

Difference of quantum statics explicitly emerges

Bose statics: BEC, superfluid $^4$He
Fermi statics: Fermi liquid, Cooper pairing

Steam, Water, Ice
Quantum Liquid

Classical Liquids

- Frozen at the absolute zero temperature
- Ordering in conventional way, e.g. crystallization

Quantum Liquids

- No freezing by quantum fluctuation
  Zero point oscillation
  > Interaction

Quantum Spin Liquids

A state of matter where strong quantum fluctuations melt the long-range magnetic order even at absolute zero temperature.
A spin pair of $S=1/2$ spins coupled by the Heisenberg interaction

$$J S_1 \cdot S_2$$

$$S_1 \cdot S_2 = S_{1x} S_{2x} + S_{1y} S_{2y} + S_{1z} S_{2z}$$

$$= \frac{1}{2} (S_{1+} S_{2-} + S_{1-} S_{2+}) + S_{1z} S_{2z}$$

**Antiferromagnetic interaction**

**Classical spin**

$$\uparrow \downarrow$$

$$< S_{1z} S_{2z} > \quad E_N = -\frac{1}{4} J$$

**Quantum spin** (no classical analogue)

$$|s> = \frac{1}{\sqrt{2}} (|\uparrow\downarrow> - |\downarrow\uparrow>)$$

**singlet**

$$< S_{1x} S_{2x} >, < S_{1y} S_{2y} >, < S_{1z} S_{2z} >$$

$$E_{sx} = E_{sy} = E_{sz} = -\frac{1}{4} J$$

**Quantum fluctuation**

$$E_s = -\frac{3}{4} J < E_N$$
1D spin-1/2 Heisenberg antiferromagnet

\[ \mathcal{H} = J \sum_{i<j}^{N} S_i \cdot S_j \]

Ground state \( S=0 \)

(1) Classical Neel state

\[ E_{Neel} = -\frac{1}{4} JN = -0.25JN \]

Spin rotational symmetry: Broken

Translational symmetry: Broken

(2) Dimer state (Valence Bond Solid State)

Spin singlet \( |s> = \frac{1}{\sqrt{2}} (|\uparrow\downarrow> - |\downarrow\uparrow>) \)

Long range order of singlet pairs

\[ E_{singlet} = -\frac{3}{8} JN = -0.375JN \]

Spin rotational symmetry: Unbroken

Translational symmetry: Broken

(3) Exact solution (Quantum Spin Liquid State)

\[ E_0 = - (\ln 2 - 1/4) JN = -0.4431JN \]

Spin rotational symmetry: Unbroken

Translational symmetry: Unbroken

1D spin-1/2 Heisenberg antiferromagnet

Ground state: Tomonaga-Luttinger liquid

**Quantum spin liquid** (No long range order)

Spin-spin correlation function: \( | <S_i \cdot S_j> | \sim |i - j|^{-1} \sim r^{-1} \)

**Algebraic spin correlation** (critical phase)

**Excitation**

Gapless (magnetic)

J. des Cloizeaux and J.J. Pearson

\[ \Delta E_1(q) = \frac{\pi}{2} J | \sin qa | \]

Spinon excitation (S=1/2, e=0)

\[ \uparrow \downarrow \uparrow \downarrow \uparrow \downarrow \uparrow \downarrow \uparrow \downarrow \]

\[ \uparrow \downarrow \uparrow \downarrow \uparrow \downarrow \uparrow \downarrow \uparrow \downarrow \]

\[ \text{No energy cost in moving the spinons far apart.} \]

Notion of QSL is firmly established in 1D. How about in higher dimensions?
QSLs in two and three dimensions

Geometrical frustrations are required

Classical  A significant ground-state degeneracy
Quantum  Quantum fluctuation lifts the degeneracy. It may lead to a QSL ground state.
A triangle of AF interacting Ising spins, all spins cannot be antiparallel.

P.W. Anderson (1973)

Quantum spin liquid state in 2D triangular lattice

Frustration

Triangle relation
What is the ground state of 2D triangular lattice?

Néel order?

Valence Bond Solid?

Spin Liquid?

\[ \frac{1}{\sqrt{2}} \left( \begin{array}{c} 1 \\ -1 \end{array} \right) \]

**Broken spin-rotational symmetry**

**Broken translational symmetry**

3-sublattice Néel order (120° structure)

**Unbroken spin-rotational**

**Broken translational**

Long range order of singlet

**Unbroken spin-rotational**

**Unbroken translational**

No simple symmetry breaking

Singlet spin pair

Broken spin-rotational symmetry

Broken lattice symmetry

Unbroken spin-rotational

Unbroken translational
Quantum spin liquid state in 2D triangular lattice

\[ \mathcal{H} = J \sum_{(i,j)=\Delta} S_i \cdot S_j \]

Heisenberg model with nearest-neighbor interaction

Resonating Valence Bond (RVB) Liquid

\[ = \frac{1}{\sqrt{2}} (| \uparrow \downarrow > - | \downarrow \uparrow >) \]

Superposition of different configurations

Resonance between highly degenerated spin configurations leads to a liquid-like wavefunction.

P. Fazekas and P.W. Anderson, Philos. Mag. (74)
Quantum spin liquid state in 2D triangular lattice

\[ \mathcal{H} = J \sum_{(i,j) = \Delta} S_i \cdot S_j \]

Heisenberg model with nearest-neighbor interaction

Resonating Valence Bond (RVB) Liquid

Superposition of different configurations

Resonance between highly degenerated spin configurations leads to a liquid-like wavefunction.

Further fluctuations, such as higher order ring exchanges, are required to realize a QSL.

3-sublattice Néel order (120° structure)

L. Caprioti, A. E.E. Trumper and S. Sorella, PRL (99)
B. Bernu, C. Lhuillier and L. Pierre, PRL (92)

P. Fazekas and P.W. Anderson, Philos. Mag. (74)
Order or not order?
• geometrical frustration plays important rule

Many theories proposed
• Resonating-valence-bond liquid
• Chiral spin liquid
• Quantum dimer liquid
• $Z_2$ spin liquid
• Algebraic spin liquid
• Spin Bose Metal
• Etc.,

Elementary excitaion not yet identified
• Spinon with Fermi surface
• Vison
• Majorana fermions

New quantum condensed-state may be realized!!
Quantum spin liquid on a 2D triangular lattice

Only a few candidate materials exist.

- Triangular lattice
- Monolayer $^3$He →
- $^3$He, $^4$He, HD/HD →
- Graphite →

$J \sim 3$ mK

Specific heat
- R. Masutomi et al., PRL 92, 025301 (04)
- K. Ishida et al. PRL 79, 3451 (97)

$^3$He on graphite
- Organic compounds

No magnetic ordering
Only a few candidate materials exist.

**Triangular lattice**

\[ \kappa-[\text{bis(ethylenedithio)tetrathiafulvalene}]_2-\text{Cu}_2-(\text{CN})_3 \]

\[ \text{C}_2\text{H}_5[\text{CH}_3]_3\text{Sb}[\text{Pd}(1,3\text{-dithiole-2-thione-4,5-dithiolate})_2]_2 \]

catechol-fused ethylenedithiotetrathiafulvalene

**Organic compounds**

\(^3\text{He on graphite}\)
Only a few candidate materials exist. 

Triangular lattice

\[ \kappa-(\text{BEDT-TTF})_2\text{Cu}_2(\text{CN})_3 \]

\[ \text{EtMe}_3\text{Sb}[\text{Pd} (\text{dmit})_2]_2 \]

\[ \kappa-\text{H}_3(\text{Cat-EDT-TTF})_2 \]
Quantum spin liquid on a 2D triangular lattice

Only a few candidate materials exist.

- Triangular lattice
- \( \kappa-(\text{BEDT-TTF})_2\text{Cu}_2(\text{CN})_3 \) \( \kappa\)-ET
- \( \text{EtMe}_3\text{Sb}[\text{Pd(dmit)}_2]_2 \) dmit
- \( \kappa-H_3(\text{Cat-EDT-TTF})_2 \) cat

\( ^3 \text{He on graphite} \)

Organic compounds
EtMe$_3$Sb[Pd(dmit)$_2$]$_2$

2D spin system

SIDE VIEW

2D layer of Pd(dmit)$_2$ molecule

Cation layer
Non-magnetic
$X = \text{EtMe}_3\text{Sb, Et}_2\text{Me}_2\text{Sb, etc.}$

S = $\frac{1}{2}$ Triangular lattice

TOP VIEW

$S_A \sim 0.5$ eV $\gg t_B, t_s, t_r$
$t_B \sim 55$ meV, $t_s \approx t_r \sim 45$ meV

Dimerization $\rightarrow$ Half-filled Mott insulator

- Very clean single crystals are available
- Many material variants can be prepared.
A new QSL system $\text{EtMe}_3\text{Sb}[\text{Pd}(\text{dmit})_2]_2$

$\beta'-(\text{Cation})[\text{Pd}(\text{dmit})_2]_2$

No magnetic order down to $\sim J/10,000$

$\chi(T)$: 2D triangular

$J = 220 \sim 250$ K

No internal magnetic field

$^{13}$C NMR

No muon spin rotation

ZF $\mu$SR

K. Kanoda and R. Kato

I. Itou et al., Nature Phys. (10)

Y. Ishii et al.
Two key questions

Elementary excitations

Gapped or gapless?
Magnetic or nonmagnetic?

Phase diagram

How the nature of the QSL varies when tuned by non-thermal parameters, such as degree of frustration?
Spin liquid

Charge order

Elementary excitations in EtMe$_3$Sb[Pd(dmit)$_2$]$_2$

\[ C_P T^{-1} = \gamma + \beta T^2 \]
\( (\gamma \approx 20 \text{ mJK}^{-2}\text{mol}^{-1}) \)

Gapless excitation

Contaminated by large Schottky contribution at low temperatures

Specific heat

Thermal conductivity

Can probe elementary excitations at low temperature very reliably.

Not affected by localized impurities
Not contaminated by Schottky contribution

\[ \frac{1}{Wt} Q = \kappa \frac{\Delta T}{\ell} \]

EtMe₃Sb[Pd(dmit)₂]₂ Spin liquid

Et₂Me₂Sb[Pd(dmit)₂]₂ Charge order

Thermal conductivity

Clear residual of $\kappa_{xx}/T$

$k_{xx}/T (T \to 0) = 0.19 \text{ W/K}^2\text{m}$

Evidence for a *gapless excitation*.

Estimation of mean free path

$C/T \sim 20\text{mJ}/\text{K}^2\text{mol}$

$\ell = 0.5\,\mu\text{m} \gg a \sim 1\,\text{nm}$

More than 500 times longer than the interspin distance!!

*Itinerant excitation*

Homogeneous

Extremely long correlation length

M. Yamashita *et al*. Science 328, 1246 (10)
Quantum spin liquid conducts heat very well, as good as brass.
Elementary excitations contain a gapless component

Next important question: Are they magnetic?

Elementary excitations in EtMe₃Sb[Pd(dmit)₂]₂

- Schematic representation of energy spectra
  - Triplet (magnetic)
  - Singlet (non-magnetic)

Type-I gapless spin liquid
- \( \Delta_s = 0, \Delta_t > 0 \)
- \( \langle S(r)S(0) \rangle \sim e^{-r/\xi} \)
  - Exponential

Type-II gapless spin liquid
- \( \Delta_s = 0, \Delta_t = 0 \)
- \( \langle S(r)S(0) \rangle \sim r^{-\nu} \)
  - Power law (algebraic)
Elementary excitations in EtMe$_3$Sb[Pd(dmit)$_2$]$_2$

**Elementary excitations contain a gapless component**

Next important question: Are they magnetic?

Uniform susceptibility and magnetization at low temperatures

SQUID (Only down to ~4 K due to large Curie contribution)

Magnetic torque+ESR (down to 30 mK up to 32 T)

\[
\tau_{c^*-a} = \frac{1}{2} \mu_0 H^2 V \Delta \chi_{c^*-a} \sin 2\theta
\]

- Torque picks up only anisotropic components
  - Isotropic contribution from impurities is cancelled.
- High sensitivity.
  - Measurements on a tiny single crystal are possible.
Uniform susceptibility and magnetization of QSL $T$-independent and remains finite at $T \to 0K$ increases linearly with $H$

Gapless excitations are magnetic (absence of spin gap)

$\Delta \propto \xi^{-1}$ ( $\xi$: magnetic correlation length, $\Delta$: spin gap )

Divergence of $\xi$, i.e. QSL is in a critical state

$\left\langle S^z(r)S^z(0) \right\rangle \propto r^{-\eta}$

Algebraic
Deuteration changes the low temperature specific heat. Presumably it reduces $J'/J$.

$J'/J \approx 0.83$ (pristine)

**h$_9$-dmit:** $C_p/T (T \to 0 \text{ K}) \approx 20 \text{ mJ/K}^2\text{mol}$

**d$_9$-dmit:** $C_p/T (T \to 0 \text{ K}) \approx 40 \text{ mJ/K}^2\text{mol}$

Deuteration changes the low temperature specific heat. Presumably it reduces $J'/J$.

Deuteration changes the degrees of geometrical frustration.
Deuteration changes the degrees of geometrical frustration. Both $h_9$- and $d_9$-dmit systems exhibit essentially the same paramagnetic behavior with gapless magnetic excitations.

Both systems are in the critical state down to $k_B T \sim J/10,000$. 
Both pristine (h₉-dmit) and deuterated (d₉-dmit) samples with different degrees of frustration exhibit essentially the same paramagnetic behavior with *gapless* magnetic excitations.

An extended quantum critical phase, rather than a QCP.
Are the excitations in the QSL fermionic or bosonic?

Fermionic excitations appear to be more likely

Gapless Nambu-Goldstone boson

Appearance of another gapless bosonic excitations seems unlikely

$k_B T \sim J$
Are the excitations in the QSL fermionic or bosonic?

A simple thermodynamic test assuming 2D Fermion with Fermi surface

**Pauli susceptibility**

\[
\chi_{\perp} = \frac{1}{4} g_e^2 \mu_B^2 D(\varepsilon_F)
\]

\[
\chi_{\perp} = 8.0(5) \times 10^{-4} \text{ emu/mol}
\]

\[
D(\varepsilon_F) = n/\varepsilon_F
\]

**Specific heat coefficient C/T**

\[
\gamma = \frac{1}{3} \pi^2 k_B^2 D(\varepsilon_F) = \frac{1}{3} \pi^2 k_B^2 \frac{4\chi_{\perp}}{g_e^2 \mu_B^2} \sim 56 \text{ mJ/K}^2 \text{ mol}
\]

\[
\gamma \sim 20 \text{ mJ/K}^2 \text{ mol} \text{ (experimental value)}
\]

**Fermi temperature**

\[
T_F = \frac{\varepsilon_F}{k_B} = \frac{g_e^2 \mu_B^2}{4\chi_{\perp} k_B} \sim 480 \text{ K} \quad J/k_B \sim 250 \text{ K} \text{ (exp. value)}
\]

**Wilson ratio**

\[
R_W = \chi_{\perp}/\gamma = 2.83(1.41) \text{ for pristine (deuterated) sample}
\]

Elementary excitations behave like Fermi liquid
A new phase in a Mott insulator

Spin excitations behave as in Pauli paramagnetic metals with Fermi surface, even though the charge degrees of freedom are frozen.

D.F. Mross and T. Senthil, PRB (11)
A new phase in a Mott insulator

**Hubbard model**

\[ \mathcal{H} = - \sum_{(i,j), \sigma} t \left( c_{i\sigma}^\dagger c_{j\sigma} + \text{H.C.} \right) + U \sum_{i=1}^{N} n_{i\uparrow} n_{i\downarrow} \]

**Non-Magnetic Insulating phase near M-I transition**

(Quantum spin liquid)

Energy resolutions of these calculations are not enough to discuss low energy excitations \((E \sim J/100)\)

- **t'/t = 1**
- **Metal**
- **NMI**
- **120° Néel**

Morita-Watanabe-Imada, JPSJ (02)

Kyung-Tremblay, PRL (06)

Yoshioka-Koga-Kawakami, PRL (10)
Question
What is the theory known to describe 2D QSL?
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• **A**: resonating-valence-bond
What is the theory known to describe 2D QSL?

• **A**: resonating-valence-bond

• **B**: chiral spin liquid
What is the theory known to describe 2D QSL?

• A: resonating-valence-bond
• B: chiral spin liquid
• C: Quantum dimer liquid
What is the theory known to describe 2D QSL?

• **A**: resonating-valence-bond
• **B**: chiral spin liquid
• **C**: Quantum dimer liquid
• **D**: QSL with spinon Fermi surface
What is the theory known to describe 2D QSL?

• **A**: resonating-valence-bond
• **B**: chiral spin liquid
• **C**: Quantum dimer liquid
• **D**: QSL with spinon Fermi surface
• **E**: Algebraic spin liquid
What is the theory known to describe 2D QSL?

- **A**: resonating-valence-bond
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- **D**: QSL with spinon Fermi surface
- **E**: Algebraic spin liquid
- **F**: $\mathbb{Z}_2$ spin liquid

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**Dynamics and Transport of the $\mathbb{Z}_2$ Spin Liquid: Application to $\kappa$-(ET)$_2$Cu$_2$(CN)$_3$**

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(Received 6 September 2008; published 29 April 2009; publisher error corrected 30 April 2009)

We describe neutron scattering, NMR relaxation, and thermal transport properties of $\mathbb{Z}_2$ spin liquids in two dimensions. Comparison to recent experiments on the spin $S = 1/2$ triangular lattice antiferromagnet in $\kappa$-(ET)$_2$Cu$_2$(CN)$_3$ shows that this compound may realize a $\mathbb{Z}_2$ spin liquid. We argue that the topological “vison” excitations dominate thermal transport, and that recent thermal conductivity experiments by M. Yamashita *et al.* have observed the vison gap.
What is the theory known to describe 2D QSL?

- **A**: resonating-valence-bond
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- **E**: Algebraic spin liquid
- **F**: $\mathbb{Z}_2$ spin liquid
- **G**: Spin-Bose-Metal phase

**Question**

Spin Bose-metal phase in a spin-$\frac{1}{2}$ model with ring exchange on a two-leg triangular strip

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(Received 4 March 2009; published 20 May 2009)

Recent experiments on triangular lattice organic Mott insulators have found evidence for a two-dimensional (2D) spin liquid in close proximity to the metal-insulator transition. A Gutzwiller wave function study of the triangular lattice Heisenberg model with a four-spin ring exchange term appropriate in this regime has found that the projected spinon Fermi sea state has a low variational energy. This wave function, together with a slave particle-gauge theory analysis, suggests that this putative spin liquid possesses spin correlations that are singular along surfaces in momentum space, i.e., “Bose surfaces.” Signatures of this state, which we will refer to as a “spin Bose metal” (SBM), are expected to manifest in quasi-one-dimensional (quasi-1D) ladder systems: the discrete transverse moments cut through the 2D Bose surface leading to a distinct pattern of 1D gapless modes. Here, we search for a quasi-1D descendant of the triangular lattice SBM state by exploring the...
What is the theory known to describe 2D QSL?

- **A**: resonating-valence-bond
- **B**: chiral spin liquid
- **C**: Quantum dimer liquid
- **D**: QSL with spinon Fermi surface
- **E**: Algebraic spin liquid
- **F**: $\mathbb{Z}_2$ spin liquid
- **G**: Spin-Bose-Metal phase
- **H**: None of the above
What is the theory known to describe 2D QSL?

- A: resonating-valence-bond
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- C: Quantum dimer liquid
- D: QSL with spinon Fermi surface
- E: Algebraic spin liquid
- F: $Z_2$ spin liquid
- G: Spin-Bose-Metal phase
- H: None of the above
What kind of spin liquid is realized in EtMe$_3$Sb[Pd(dmit)$_2$]$_2$?

**Spin liquid with spinon Fermi surface**

**Gapless Spin Liquid**

**Resonating-Valence-Bond theory**

**Spin Bose Metal**

**Algebraic spin liquid**

**Gapless Spin Liquid**

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**Spin Bose-metal phase in a spin-$\frac{1}{2}$ model with ring exchange on a two-leg triangular strip**

D. N. Sheng,$^{1}$ Olexei I. Motrunich,$^{2}$ and Matthew P. A. Fisher$^{3}$  

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**Quantum orders and symmetric spin liquids**

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A concept—quantum order—is introduced to describe a new kind of orders that generally appear in quantum states at zero temperature. Quantum orders that characterize the universality classes of quantum states described by complex ground-state wave functions are much richer than classical orders that characterize the universality classes of finite-temperature classical states (described by positive probability distribution functions). Landau’s theory for orders and phase transitions does not apply to quantum orders since they cannot be described by broken symmetries and the associated order parameters. We introduce a mathematical object—projective symmetry group—to characterize quantum orders. With the help of quantum orders and projective symmetry groups, we construct hundreds of symmetric spin liquids, which have SU(2), U(1), or Z$_2$ gauge

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**Strong Mott insulators on the triangular lattice: Possibility of a gapless nematic quantum spin liquid**

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We study the energetics of Gutzwiller projected BCS states of various symmetries for the triangular lattice antiferromagnet with a four-particle ring exchange using variational Monte Carlo methods. In a range of parameters the energetically favored state is found to be a projected $d_{x^2-y^2}$-paired state which breaks lattice rotational symmetry. We show that the properties of this nematic or orientationally ordered paired spin liquid state as a function of temperature and pressure can account for many of the experiments on organic materials. We also study the ring-exchange model with ferromagnetic Heisenberg exchange and find that among the studied ansätze, a projected $f$-wave state is the most favorable.

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**Gapless Spin Liquids: Stability and Possible Experimental Relevance**

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(Dated: September 3, 2012)

For certain crystalline systems, most notably the organic compound EtMe$_3$Sb[Pd(dmit)$_2$]$_2$, experimental evidence has accumulated of an insulting state with a high density of gapless neutral excitations that produce Fermi-liquid-like power laws in thermodynamic quantities and thermal transport. This has been taken as evidence of a fractionalized spin liquid state. In this paper, we argue that if the experiments are taken at face value, the most promising spin liquid candidates are a Z$_2$ spin liquid with a pseudo-Fermi surface and no broken symmetries, or a Z$_2$ spin liquid with a pseudo-Fermi surface and at least one of the following spontaneously broken: (a) time-reversal and (b) spin-charge-conjugation symmetry.
Thermal Hall effect

\[ \tan \theta_H = \frac{k_{xy}}{k_{xx}} \]

H. Katsura, N. Nagaosa and P.A. Lee, PRL (10)

No discernible thermal Hall effect

The coupling between the magnetic field and the gauge flux may be weak.

Quantum oscillation

O.I. Mitrunch, PRB (06)

No discernible oscillation

M. Yamashita et al., Science (10)

Remaining questions
Elementary excitations and phase diagram of the QSL in $\text{EtMe}_3\text{Sb}[\text{Pd(dmit)}_2]_2$

1. Presence of gapless magnetic excitations.
2. Quantum critical phase, rather than a quantum critical point
3. Character of the excitations: likely to be fermionic

Spin excitations behave as in Pauli paramagnetic metals with Fermi surface, even though the charge degrees of freedom are frozen.

A novel Pauli paramagnetic phase in the Mott insulator

M. Yamashita et al., Science 328, 1246 (10).
D. Watanabe et al., Nature Commun. 3, 1090 (12).