Detecting collective excitations of quantum spin liquids









Collective excitations of quantum matter

- Fermi liquid zero sound and paramagnons
- Superfluid phonons and vortices
- Quantum hall liquids magnetoplasmons
- Antiferromagnets spin waves

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- Spin liquids visons and "photons"

Antiferromagnet















General approach

Look for spin liquids across continuous (or weakly first-order) quantum transitions from antiferromagnetically ordered states

Outline

- I. Collective excitations of spin liquids in two dimensions Photons and visons
- 2. Detecting the vison Thermal conductivity of κ -(ET)₂Cu₂(CN)₃
- 3. Detecting the photon Valence bond solid order around Zn impurities

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 I. Collective excitations of spin liquids in two dimensions
 Photons and visons

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- **3. Detecting the photon** *Valence bond solid order around Zn impurities*

Square lattice antiferromagnet



Ground state has long-range Néel order

Order parameter is a single vector field $\vec{\varphi} = \eta_i \vec{S}_i$ $\eta_i = \pm 1$ on two sublattices $\langle \vec{\varphi} \rangle \neq 0$ in Néel state. Square lattice antiferromagnet



Destroy Neel order by perturbations which preserve full square lattice symmetry

A.W. Sandvik, *Phys. Rev. Lett.* **98**, 2272020 (2007). R.G. Melko and R.K. Kaul, *Phys. Rev. Lett.* **100**, 017203 (2008).

Theory for loss of Neel order

Write the spin operator in terms of Schwinger bosons (spinons) $z_{i\alpha}$, $\alpha = \uparrow, \downarrow$:

$$\vec{S}_i = z_{i\alpha}^{\dagger} \vec{\sigma}_{\alpha\beta} z_{i\beta}$$

where $\vec{\sigma}$ are Pauli matrices, and the bosons obey the local constraint

$$\sum_{\alpha} z_{i\alpha}^{\dagger} z_{i\alpha} = 2S$$

Effective theory for spinons must be invariant under the U(1) gauge transformation

$$z_{ilpha}
ightarrow e^{i heta} z_{ilpha}$$

Perturbation theory

Low energy spinon theory for "quantum disordering" the Néel state is the ${\rm CP^1}$ model

$$S_{z} = \int d^{2}x d\tau \left[c^{2} \left| \left(\nabla_{x} - iA_{x} \right) z_{\alpha} \right|^{2} + \left| \left(\partial_{\tau} - iA_{\tau} \right) z_{\alpha} \right|^{2} + s \left| z_{\alpha} \right|^{2} \right. \right. \\ \left. + u \left(\left| z_{\alpha} \right|^{2} \right)^{2} + \frac{1}{4e^{2}} (\epsilon_{\mu\nu\lambda} \partial_{\nu} A_{\lambda})^{2} \right]$$

where A_{μ} is an emergent U(1) gauge field (the "**photon**") which describes low-lying spin-singlet excitations.

Phases:

$$\langle z_{\alpha} \rangle \neq 0 \qquad \Rightarrow \qquad \text{N\'eel (Higgs) state}$$

 $\langle z_{\alpha} \rangle = 0 \qquad \Rightarrow \qquad \text{Spin liquid (Coulomb) state}$





N. Read and S. Sachdev, *Phys. Rev. Lett.* **62**, 1694 (1989)

From the square to the triangular lattice



A spin density wave with

 $\langle \vec{S}_i \rangle \propto (\cos(\mathbf{K} \cdot \mathbf{r}_i, \sin(\mathbf{K} \cdot \mathbf{r}_i)))$

and $\mathbf{K} = (\pi, \pi)$.

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A spin density wave with

 $\langle \vec{S}_i \rangle \propto (\cos(\mathbf{K} \cdot \mathbf{r}_i, \sin(\mathbf{K} \cdot \mathbf{r}_i)))$

and $\mathbf{K} = (\pi + \Phi, \pi + \Phi)$.

Interpretation of non-collinearity Φ

Its physical interpretation becomes clear from the allowed coupling to the spinons:

$$S_{z,\Phi} = \int d^2 r d\tau \left[\lambda \Phi^* \epsilon_{\alpha\beta} z_{\alpha} \partial_x z_{\beta} + \text{c.c.} \right]$$

 Φ is a spinon pair field

N. Read and S. Sachdev, Phys. Rev. Lett. 66, 1773 (1991)



Spin liquid with a **"photon"** collective mode [Unstable to valence bond solid (VBS) order]

$$\langle z_{\alpha} \rangle = 0$$

S



non-collinear Néel state

 S_{C}

N. Read and S. Sachdev, *Phys. Rev. Lett.* **66**, 1773 (1991)

 Z_2 spin liquid with a

 $\langle z_{\alpha} \rangle = 0 , \ \langle \Phi \rangle \neq 0$

S

vison excitation

What is a vison ?

A vison is an Abrikosov vortex in the spinon pair field Φ .

In the Z_2 spin liquid, the vison is S = 0quasiparticle with a finite energy gap

N. Read and S. Sachdev, Phys. Rev. Lett. 66, 1773 (1991)





























Global phase diagram



S. Sachdev and N. Read, Int. J. Mod. Phys B 5, 219 (1991), cond-mat/0402109

Mutual Chern-Simons Theory

Express theory in terms of the physical excitations: the spinons, z_{α} , and the visons. After accounting for Berry phase effects, the visons can be described by a complex field v, which transforms non-trivially under the square lattice space group operations.

The spinons and visons have mutual semionic statistics, and this leads to the continuum theory:

$$S = \int d^2x d\tau \left[c^2 \left| (\nabla_x - iA_x) z_\alpha \right|^2 + \left| (\partial_\tau - iA_\tau) z_\alpha \right|^2 + s \left| z_\alpha \right|^2 + \dots \right. \\ \left. + \widetilde{c}^2 \left| (\nabla_x - iB_x) v \right|^2 + \left| (\partial_\tau - iB_\tau) v \right|^2 + \widetilde{s} \left| v \right|^2 + \dots \right. \\ \left. + \frac{i}{\pi} \epsilon_{\mu\nu\lambda} B_\mu \partial_\nu A_\lambda \right]$$

Cenke Xu and S. Sachdev, to appear

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This theory fully accounts for all the phases, including their global topological properties and their broken symmetries. It also completely describe the quantum phase transitions between them.
Global phase diagram



Cenke Xu and S. Sachdev, to appear

<u>Mutual Chern-Simons Theory</u>

$$S = \int d^2x d\tau \left[c^2 \left| (\nabla_x - iA_x) z_\alpha \right|^2 + \left| (\partial_\tau - iA_\tau) z_\alpha \right|^2 + s \left| z_\alpha \right|^2 + \dots \right]$$

 $+ \tilde{c}^{2} \left| (\nabla_{x} - iB_{x})v \right|^{2} + \left| (\partial_{\tau} - iB_{\tau})v \right|^{2} + \tilde{s} \left| v \right|^{2} + \ldots + \frac{i}{\pi} \epsilon_{\mu\nu\lambda} B_{\mu} \partial_{\nu} A_{\lambda} \right|$

Low energy states on a torus:

Г

- Z_2 spin liquid has a 4-fold degeneracy.
- Non-collinear Néel state has low-lying tower of states described by a broken symmetry with order parameter S_3/Z_2 .
- "Photon" spin liquid has a low-lying tower of states described by a broken symmetry with order parameter S_1/Z_2 . This is the VBS order ~ v^2 .
- Néel state has a low-lying tower of states described by a broken symmetry with order parameter $S_3 \times S_1/(U(1) \times U(1)) \equiv S_2$. This is the usual vector Néel order.

Cenke Xu and S. Sachdev, to appear

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2. Detecting the vison *Thermal conductivity of* κ -(*ET*)₂*Cu*₂(*CN*)₃

3. Detecting the photon Valence bond solid order around Zn impurities The following slides and thermal conductivity data are from:

M. Yamashita, H. Nakata, Y. Kasahara, S. Fujimoto, T. Shibauchi, Y. Matsuda, T. Sasaki, N. Yoneyama, and N. Kobayashi, preprint and 25th International Conference on Low Temperature Physics, SaM3-2, Amsterdam, August 9, 2008.

Q2D organics κ-(ET)₂X; spin-1/2 on triangular lattice



κ -(BEDT-TTF)₂Cu₂(CN)₃



•face-to-face pairs of BEDT-TTF molecules form dimers by strong coupling.

•Dimers locate on a vertex of triangular lattice and ratio of the transfer integral is ~1.

 $t'_t = 1.06, U_t = 8.2$

•Charge +1 for each ET dimer; Half-filling Mott insulator.



•Cu₂[N(CN)₂]Cl (t'/t = 0.75, U/t = 7.8) Néel order at T_N =27 K

•Cu₂(CN)₃ (t'/t = 1.06. U/t = 8.2) No sign of magnetic order down to 1.9 K.

Heisenberg High-T Expansion (PRL, **71** 1629 (1993)) <u>J ~ 250 K</u>

Y. Shimizu etal, PRL 91, 107001 (2003)

Spin excitation in κ - $(ET)_2Cu_2(CN)_3$

¹³C NMR relaxation rate

Shimizu et al., PRB 70 (2006) 060510



 $1/T_1 \sim \text{power law of T}$

Low-lying spin excitation at low-T



Heat capacity measurements

Thermodynamic properties of a spin-1/2 spin-liquid state in a κ -type organic salt

$$\gamma = 15 \frac{\text{mJ}}{\text{K}^2 \text{mol}}$$

Evidence for Gapless spinon?





S. Yamashita, et al., Nature Physics **4**, 459 - 462 (2008)

A. P. Ramirez, Nature Physics 4, 442 (2008)

Thermal-Transport Measurements

Only itinerant excitations carrying entropy can be measured without localized ones

- no impurity contamination

 - 1/T₁, χ measurement ← free spins
 Heat capacity ← Schottky contamination

Best probe to reveal the low-lying excitation at low temperatures.

$$\kappa/T \qquad \gamma = 0? \neq 0? \qquad \qquad \frac{\kappa}{T} \approx \frac{1}{T} (C \approx \varkappa) \propto \gamma + \beta T^{2} \qquad \begin{cases} C \propto \gamma T + \beta T^{3} \\ \gamma : \text{Gapless spin liquid} \\ (\text{Spinon}) \\ \beta : \text{Phonon} \end{cases}$$

•What is the low-lying excitation of the quantum spin liquid found in κ -(BEDT-TTF)₂Cu₂(CN)₃.
•Gapped or Gapless spin liquid? Spinon with a Fermi surface?

Thermal Conductivity below 10K



Thermal Conductivity below 300 mK



Arrhenius plot $\kappa = \alpha \exp\left(-\frac{\Delta}{k_B T}\right) + \beta T^3$ 10^{-1} •Arrhenius behavior for $T < \Delta$! 6 •Tiny gap 4 ≻ ∆ = 0.46 K ~ J/500 $\begin{bmatrix} & & & & \\$ 2 8 6 H = 0 Tesla 4 $\alpha = 0.12 \text{ W} \text{ sc}^{-1} \text{ sm}$ $\Delta = 0.46 \, \mathrm{K}$ 2 $\beta = 0.51 \,\mathrm{W}\,\mathrm{xK}^{-4}\,\mathrm{xm}^{-1}$ 10⁻³ 12 4 6 8 10 14 1/T(1/K)

I. Thermal conductivity is dominated by vison transport

The thermal conductivity (per layer) of N_v species of slowly moving visons of mass m_v , above an energy gap Δ_v , scattering off impurities of density $n_{\rm imp}$ is

$$\kappa_v = \frac{N_v m_v k_B^3 T^2 \ln^2 (T_v/T) e^{-\Delta_v/(k_B T)}}{4\pi \hbar^3 n_{\rm imp}}.$$

where T_v is of order the vison bandwidth.

Yang Qi, Cenke Xu and S. Sachdev, arXiv:0809:0694

I. Thermal conductivity is dominated by vison transport



Best fit to data yields, $\Delta_v \approx 0.24$ K and $T_v \approx 8$ K.

Yang Qi, Cenke Xu and S. Sachdev, arXiv:0809:0694

Thermal Conductivity below 10K



Spin excitation in κ - $(ET)_2Cu_2(CN)_3$

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Shimizu et al., PRB 70 (2006) 060510



 $1/T_1 \sim \text{power law of T}$

Low-lying spin excitation at low-T



II. NMR relaxation is caused by spinons close to the quantum critical point between the non-collinear Néel state and the Z_2 spin liquid

The quantum-critical region of magnetic ordering on the triangular lattice is described by the O(4) model and has

$$\frac{1}{T_1} \sim T^{\bar{\eta}}$$

where $\bar{\eta} = 1.374(12)$. This compares well with the observed $1/T_1 \approx T^{3/2}$ behavior.

(Note that the singlet gap is associated with a spinon-pair excitations, which is distinct from the vison gap, Δ_v .) Yang Qi, Cenke Xu and S. Sachdev, arXiv:0809:0694

Global phase diagram



Cenke Xu and S. Sachdev, to appear

II. NMR relaxation is caused by spinons close to the quantum critical point between the non-collinear Néel state and the Z_2 spin liquid

At higher $T > \Delta_v$, the NMR will be controlled by the spinonvison multicritical point, described by the mutual Chern-Simons theory. This multicritical point has

$$\frac{1}{T_1} \sim T^{\eta_{\rm cs}}$$

We do not know the value η_{cs} accurately, but the 1/N expansion (and physical arguments) show that $\eta_{cs} < \overline{\eta}$.

Spin excitation in κ - $(ET)_2Cu_2(CN)_3$

¹³C NMR relaxation rate

Shimizu et al., PRB 70 (2006) 060510



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Low-lying spin excitation at low-T



III. The thermal conductivity of the visons is larger than the thermal conductivity of spinons

We compute the spinon thermal conductivity (κ_z) by the methods of quantum-critical hydrodynamics (developed recently using the AdS/CFT correspondence). Because the spinon bandwidth $(T_z \sim J \sim 250 \text{ K})$ is much larger than the vison mass/bandwidth $(T_v \sim 8 \text{ K})$, the vison thermal conductivity is much larger over the T range of the experiments.



Yang Qi, Cenke Xu and S. Sachdev, arXiv:0809:0694

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Non-perturbative effects in U(1) spin liquid

N. Read and S. Sachdev, *Phys. Rev. Lett.* **62**, 1694 (1990) T. Senthil, A. Vishwanath, L. Balents, S. Sachdev and M.P.A. Fisher, *Science* **303**, 1490 (2004).

Non-perturbative effects in U(1) spin liquid

• Monopole proliferation leads to an energy gap of the photon mode, but this gap is very small not too far from the transition to the Néel state.

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- Monopole proliferation leads to an energy gap of the photon mode, but this gap is very small not too far from the transition to the Néel state.
- Due to monopole Berry phases, the spin liquid state is unstable to valence bond solid (VBS) order, characterized by a complex order parameter $\Psi_{\rm vbs}$.

N. Read and S. Sachdev, *Phys. Rev. Lett.* **62**, 1694 (1990) T. Senthil, A. Vishwanath, L. Balents, S. Sachdev and M.P.A. Fisher, *Science* **303**, 1490 (2004).



$$\Psi_{\rm vbs}(i) = \sum_{\langle ij \rangle} \vec{S}_i \cdot \vec{S}_j e^{i \arctan(\mathbf{r}_j - \mathbf{r}_i)}$$



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Non-perturbative effects in U(1) spin liquid

- Monopole proliferation leads to an energy gap of the photon mode, but this gap is very small not too far from the transition to the Néel state.
- Due to monopole Berry phases, the spin liquid state is unstable to valence bond solid (VBS) order, characterized by a complex order parameter $\Psi_{\rm vbs}$.
- The (nearly) gapless photon is the Goldstone mode associated with the emergent circular symmetry

$$\Psi_{\rm vbs} \to \Psi_{\rm vbs} e^{i}$$
N. Read and S. S

N. Read and S. Sachdev, *Phys. Rev. Lett.* **62**, 1694 (1990) T. Senthil, A. Vishwanath, L. Balents, S. Sachdev and M.P.A. Fisher, *Science* **303**, 1490 (2004).

$$\mathcal{H}_{\mathrm{SU}(2)} = J \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j - Q \sum_{\langle ijkl \rangle} \left(\mathbf{S}_i \cdot \mathbf{S}_j - \frac{1}{4} \right) \left(\mathbf{S}_k \cdot \mathbf{S}_l - \frac{1}{4} \right)$$

Quantum Monte Carlo simulations display convincing evidence for a transition from a

Neel state at small Q to a VBS state at large Q

A.W. Sandvik, *Phys. Rev. Lett.* 98, 2272020 (2007).
R.G. Melko and R.K. Kaul, *Phys. Rev. Lett.* 100, 017203 (2008).
F.-J. Jiang, M. Nyfeler, S. Chandrasekharan, and U.-J. Wiese, arXiv:0710.3926

$$\mathcal{H}_{\mathrm{SU}(2)} = J \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j - Q \sum_{\langle ijkl \rangle} \left(\mathbf{S}_i \cdot \mathbf{S}_j - \frac{1}{4} \right) \left(\mathbf{S}_k \cdot \mathbf{S}_l - \frac{1}{4} \right)$$
$$\left| \mathrm{Im} [\Psi_{\mathrm{vbs}} \right]$$



Distribution of VBS order Ψ_{vbs} at large Q

 $\operatorname{Re}[\Psi_{vbs}]$

Emergent circular symmetry is evidence for U(1) photon and topological order

A.W. Sandvik, Phys. Rev. Lett. 98, 2272020 (2007).

Non-magnetic (Zn) impurity in the U(1) spin liquid

• The dominant perturbation of the impurity is a "Wilson line" in the time direction

$$S_{\rm imp} = i \int d\tau A_{\tau}(\mathbf{r} = 0, \tau)$$

A. Kolezhuk, S. Sachdev, R. R. Biswas, and P. Chen, Phys. Rev. B 74, 165114 (2006).

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• The dominant perturbation of the impurity is a "Wilson line" in the time direction

$$S_{\rm imp} = i \int d\tau A_{\tau}(\mathbf{r} = 0, \tau)$$

• Upon approaching the impurity the VBS (monopole) operator has a vortex-like winding in its OPE:

$$\lim_{\mathbf{r}\to 0} \Psi_{\rm vbs}(\mathbf{r},\tau) \sim |\mathbf{r}|^{\alpha} e^{i\theta}$$

where $\theta = \arctan(y/x)$.

M. A. Metlitski and S. Sachdev, Phys. Rev. B 77, 054411 (2008).





















Bond order from QMC of J-Q model



R. K. Kaul, R. G. Melko, M. A. Metlitski and S. Sachdev, arXiv:0808.0495

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R. K. Kaul, R. G. Melko, M. A. Metlitski and S. Sachdev, arXiv:0808.0495

<u>Conclusions</u>

 Vison and spinon excitations of a Z₂ spin liquid: possible explanation for NMR and thermal conductivity measurements on K-(ET)₂Cu₂(CN)₃

 Signature of photon in circular distribution of VBS order around Zn impurity: possibly relevant for STM studies of bond order in underdoped cuprates.

