

Title: Discovery of an ultra-quantum spin-liquid

Speakers: Chandra Varma

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Abstract: I will talk on experiments and their interpretation done with Professor Lei Shu and her collaborators at Fudan University, Shanghai, and some tentative theory for the observations. Thermodynamic and magnetic relaxation measurements in zero and finite magnetic field have been performed in two related almost triangular lattices of $S=1/2$ spins. One of these compounds is the purest of any of the potential spin-liquid compounds investigated so far. All its measured properties are extra-ordinary and characterized simply by just one parameter, the exchange energy obtained from susceptibility measurements. There are also colossal ultra-low energy singlet excitations. This may be the first characterization of the intrinsic properties of a class of spin-liquids. An ansatz in which the excitations are calculated from a state of singlet-dimers interacting with excitations from other such singlets can be expressed in terms of Majoranas and gives properties similar to those observed.



Discovery of an ultra-quantum spin-liquid
and a new ansatz for spin-liquids.

Chandra Varma

Talk at Perimeter Institute, May 3, 2021

Experiments:

Discovery of an ultra-quantum spin-liquid

arXiv:2102.09271

Y. X. Yang, C. Tan, Z. H. Zhu, J. Zhang, Z. F. Ding, Q. Wu, C. S. Chen T. Shiroka,
D. E. MacLaughlin, C. M. Varma, L. Shu

Spin-liquids are an example of what happens to a field when theory is done without firm experimental data to serve as a guide and a filter.

Experiments that I will talk about reveal the intrinsic behavior of a class of spin-liquids.
One can characterize its low energy scale-invariant fluctuations.
Also colossal ultra-low energy singlet fluctuations.

A new ansatz for spin-liquids suggested by the experiments.
Theory done with H.R. Krishnamurthy - IISc, Bangalore.



Structure

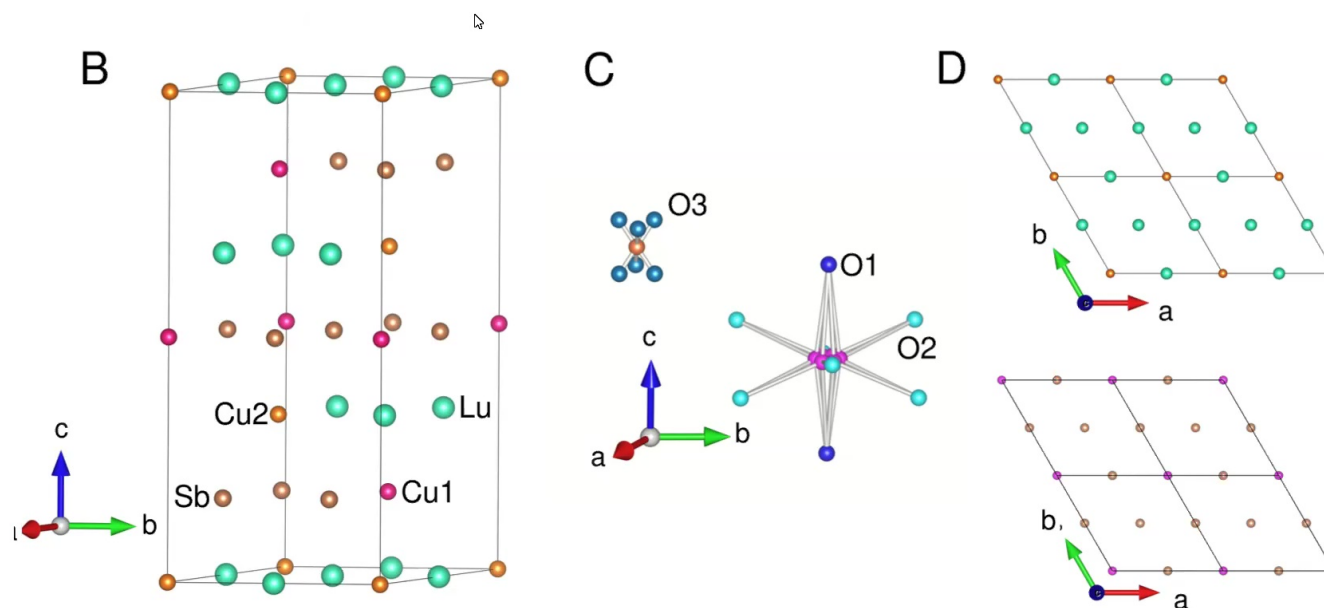


Kagome layers of Lu and of Sb, with Zn or Cu in hexagonal sites of the Kagome forming (triangular lattices with up to 3% distortions.) :

$Lu_3Zn_2Sb_3O_{14}$: Non-magnetic

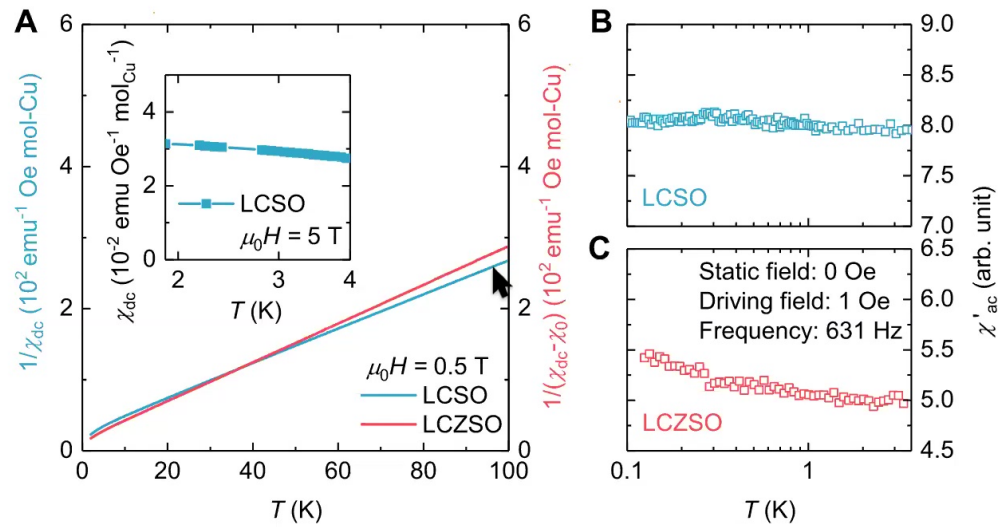
$Lu_3Cu_2Sb_3O_{14}$

$Lu_3ZnCuSb_3O_{14}$



Detailed Hamiltonian not know but is it magnetically two-dimensional?

Uniform Magnetic Susceptibility



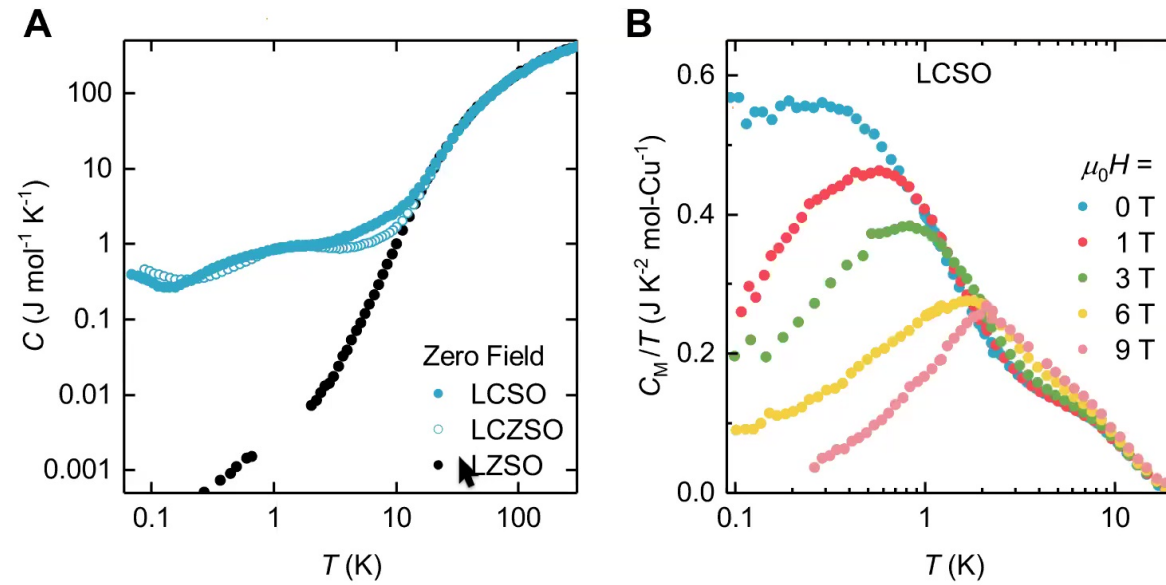
In LSCO, "orphan" spins $< 10^{-3}$, about 1% Schottky impurities

$$\theta_{W1} \approx 4.4K$$

$$\theta_{W2} \approx 26K$$



Measured specific heat and deduced Magnetic specific heat.

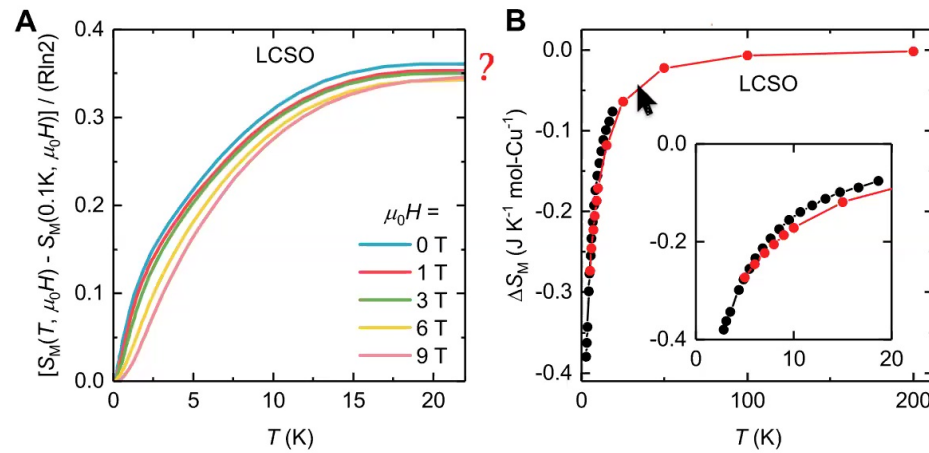


C_M in LCSO from subtraction of LZSO, a Schottky ($n = 0.014$), and a nuclear sp. heats

$C_M/T = \text{constant}$ does not by itself imply a Fermi-surface.



Available Magnetic Entropy

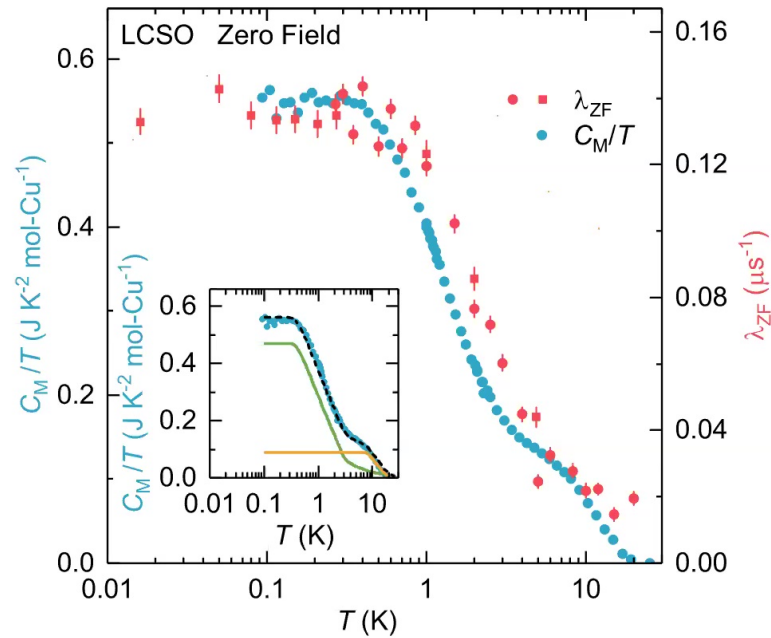


$$\Delta S_M(T) = S_M(9 \text{ T}, T) - S_M(0 \text{ T}, T)$$

from $dM/dT(H)$

Missing Entropy $\approx 57 \%$
Independent of mag. fld up to 9 Tesla.

μ -relaxation rate and $C_M(T)/T$.



1. $\lambda(T) \propto C_M(T)/T$, constant at low T. Unlike for excitations across a Fermi-surface.
2. $C_M(T)/T$ separable into two parts with equal S,
Ratio of Low T Values $\approx \theta_{W1}/\theta_{W2} \rightarrow$ Weakly Interacting layers.
3. Decay at higher T as $\ln(\theta_W/T)$
4. From $\lambda \approx 2\pi \langle H_{lc}^2 \rangle \tau$,
 $1/\tau \approx 10K \approx \theta_W$.

Summary of Experimental Results with a theoretical perspective.



1. Every measured property is characterized quantitatively by the same one parameter - an exchange energy.
2. Interesting phenomena start as $\ln(\theta_W/T)$ and follows from scale invariant fluctuations.
3. Specifically a fluctuation spectra

$$\chi''(\omega, T) = c \frac{\mu_B^2}{\theta_W} \tanh\left(\frac{\omega}{2T}\right)$$

is required, (with a logarithmic cut-off) i.e. spin-correlations $\sim 1/\tau$, in the measured temperature range, which is down to $10^{-3}\theta_W$.

4. c is required for ultra-low energy or ultra-quantum SINGLET fluctuations, which have (coincidentally?) the entropy of dimers on a triangular lattice.

Ultra-quantum fluctuations do not respond to field in the range applied.



Two 'related' compounds.

Herbertsmithite ($S=1/2$ on a Kagome lattice) has a contribution $C/T = \text{constant}$ at low T but only on applying H , and a non-magnetic analog is not available to deduce if there is unmeasurable entropy, and the compound has about 5% Schottky impurities, like our LCZSO.

Great virtue: Single crystals are available and inelastic neutron scattering showing scale-invariant flucs. but not the simplest I expect in LCSO.

YbMgO_4 : (triangular ' $s=1/2$ ') Similar thermal properties (after re-analysis) but all entropy recovered.

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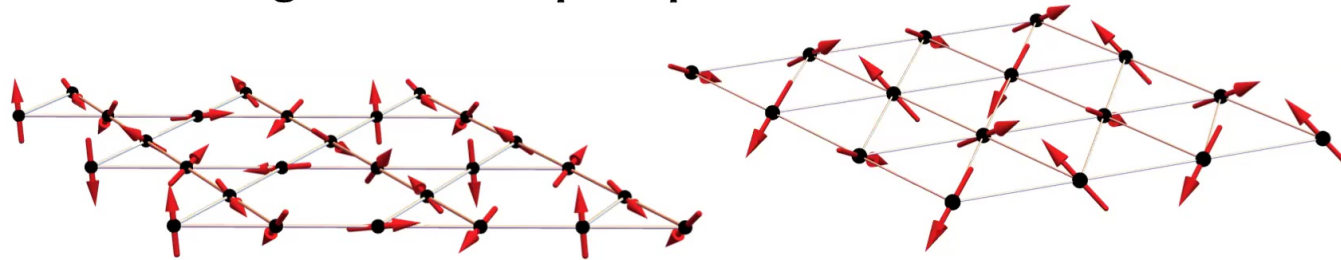
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A new ansatz for
 $S=1/2$ Heisenberg model on a triangular or Kagome lattice.

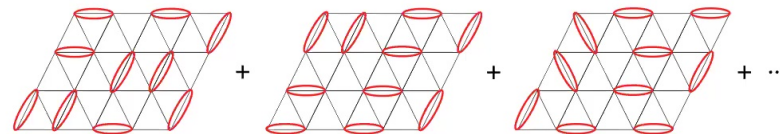
$$H = J \sum_{i,j(ni)} S_i \cdot S_j + \dots$$

Best current belief: triangular lattice - non co-planar order,
liable to be unstable to small perturbations.
Kagome lattice: Spin-liquid. What kind?




Early desperate days: RVB - Partons, $SU(2)$, $U(1)$, Z_2, \dots

Quantum liquid of dimers - Rokhsar-Kivelson, Moessner-Sondhi



A New Ansatz

Given the dark entropy, a systematic dimer based approach is worth investigating, but with actual couplings. 

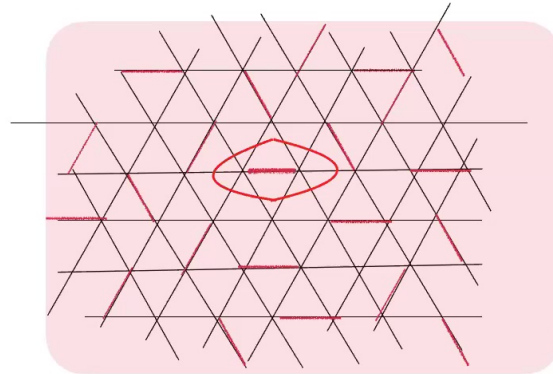
$$H = \sum_{\mu} H_{\mu} = H_{\mu 0} + H'_{\mu},$$

μ - the centers of a given configuration of dimers,

$$H_{\mu 0} = J S_{\mu \ell} \cdot S_{\mu r},$$

$$H'_{\mu} = J \frac{1}{z} \sum_{\nu(n\mu)} S_{\mu \ell} \cdot S_{\nu r} + S_{\mu r} \cdot S_{\nu \ell}$$

Try to solve the problem of a dimer interacting self-consistently with the interacting dimers.



First step - a dimer interacting with spin-flucs. with constant density of states.





First step on a related problem solved by Wilson numerical RG (B. Jones, V -1988) and by analytical methods, Affleck and Ludwig (1991), (Sire, V, Krishnamurthy -1992).

Two- Interacting Kondo impurities:

$$\begin{aligned}
 H &= K S_\ell \cdot S_r + \frac{J}{2} (S_\ell + S_r) \cdot \psi_{\text{even}}^+ \sigma \psi_{\text{even}} \\
 &+ \frac{J}{2} (S_\ell - S_r) \cdot \psi_{\text{odd}}^+ \sigma \psi_{\text{odd}} \\
 &+ K.E. (\text{even and odd fermions}).
 \end{aligned}$$

4 families of one-dimensional fermions. Only the odd parity spin-flip channel is relevant. Also express S's in terms of fermions with same number of degrees of freedom and correct commutations. (2 site version of J-W transform.)*

$$S_{\ell,r}^z = d_{\ell,r}^+ d_{\ell,r} - 1/2; S_{\ell,r}^- = d_{\ell,r} (1 - (1 \mp i) n_{r,\ell}).$$

Problem reduces in strong coupling limit to 8 state problem + K.E.

$$\begin{aligned}
 H &\rightarrow h(\{n_r, n_\ell, n_{c,0}\} = \{0, 1\}) \\
 &+ t(c_0^+ c_1 + \dots) + H.C.
 \end{aligned}$$

.....
 *The troubles with $S_i \rightarrow \psi_{i,\alpha}^+ \sigma \psi_{i,\beta}$: 2 degrees of freedom changed to 4, and Gauge redundancy. Much confusion let loose.

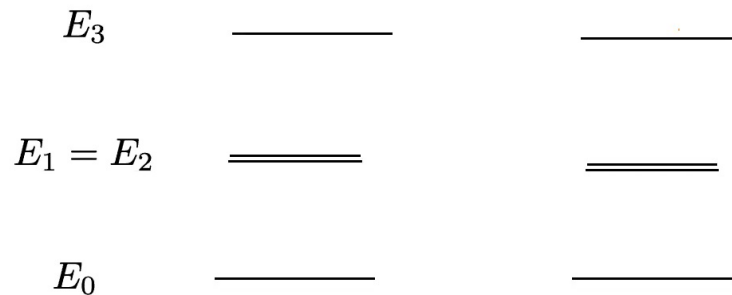


8 state problem which is two independent 4 state problems.

(Sire, V., Krishnamurthy -1992)

Basis: $(0,0,0), (0,1,1), (1,0,1), (1,1,0); (1,1,1), (1,0,0), (0,1,0), (0,0,1)$.

Degeneracy of Even charge & Odd charge states : Super -symmetry.



Odd and even sectors are coupled by c_0, c_0^+

At a critical value $J/K = r_c$

All $\langle f|c_0|i \rangle = \langle f|c_0^+|i \rangle$.

$c_0 = c_0^+$ are real fermion, i.e. they are Majoranas and may be represented by Ising spins.

This property is preserved order by order for all $c_n = c_n^+$

Equivalent to CFT (Affleck-Ludwig, Maldacena-Ludwig): $SO(7) \times$ Ising.



A mean-field ansatz keeping the symmetries:

Strong-coupling (as well as the CFT solution) involves complicated many-body operator.

Essentially the same answers obtained from a mean-field theory keeping the symmetries of the original problem. "Axial-charge" Symm.

The same thermodynamics and corr. function obtained as exact solution.

This ansatz can be extended to the lattice.

Mean-field theory for dimer interacting with a bath of fermions:

$$H_{eff} = m(d_{\mu,l}^+ d_{\mu,r} + d_{\mu,r}^+ d_{\mu,l}) + ih(d_{\mu,l}^+ d_{\mu,r}^+ + d_{\mu,l} d_{\mu,r}) \\ + c(J_1(d_{\mu,l}^+ - d_{\mu,r}) + iJ_2(d_{\mu,l} - d_{\mu,r}^+)) + H.C.$$

m, h, J_1, J_2 are self-consistently determined coeffs.

Solution: At $J_1 = 0$, or $J_2 = 0$, or $J_1 = J_2$, choose $m = h$. Then c is a majorana.

The same thermodynamics and corr. function obtained as exact solution for first two choices.

Lattice:

A Majorana can be bi-linear only with another Majorana.

So, the lattice problem at criticality is an effective Hamiltonian with four degrees of freedom per dimer, two of which are Majoranas, which can be written in terms of the d-operators at every dimer.





Mean-field ansatz for the lattice:

$$H = H_0 + H_1,$$

Intra-dimer- two states/dimer: $H_0 = \sum_{\mu} m d_{\mu l}^+ d_{\mu r} + i h d_{\mu l}^+ d_{\mu r}^+ + H.C.$

Inter-dimer-two Majoranas per dimer
with known relation of γ 's and d 's $H_1 = \frac{J}{z} \sum'_{\nu(n.\mu)} \gamma_{\mu} \tilde{\gamma}_{\nu}.$

μ, ν lie on one of the arrangements of dimers on the triang. lattice.

Simple quadratic problem except that

Assume it is not a periodic arrangement. Then, can calculate only local freq. dep. Green's functions and correlations.

With a constant local density of states of Majoranas, the results at $m=h$ are



Properties:

1. Local freq. dep. Green's functions:

$$\langle d_\ell d_\ell^+ \rangle, \langle d_\ell d_\ell \rangle, \langle d_\ell d_r \rangle, \langle d_\ell d_r^+ \rangle$$

all have a simple pole at $\omega = 0$ and a continuum with a scale $\Gamma = \rho J^2$. This gives a

$$\chi''(\omega) \propto \tanh(\beta\omega/2), \text{ for } \omega \ll \Gamma. \text{ Prediction for neutron scattering.}$$

2. $C/T \propto \lambda(T) \propto \text{constant}$, follow.

3. Entropy down to very low scale to arrangements

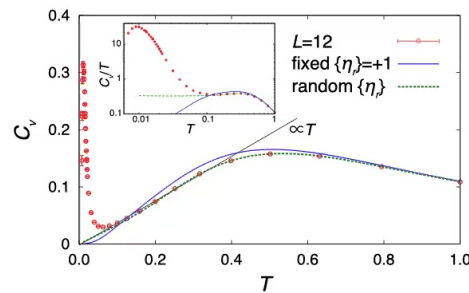
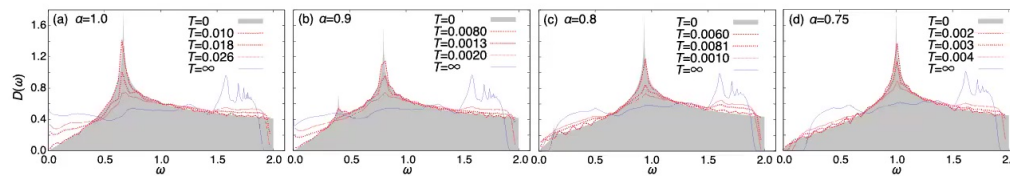
~ of dimers: 53%. Obviously spin-singlets

4. Uniform susceptibility regular, Staggered susceptibility - log T divergence.

Models with reliable similar results but no apparent connection.



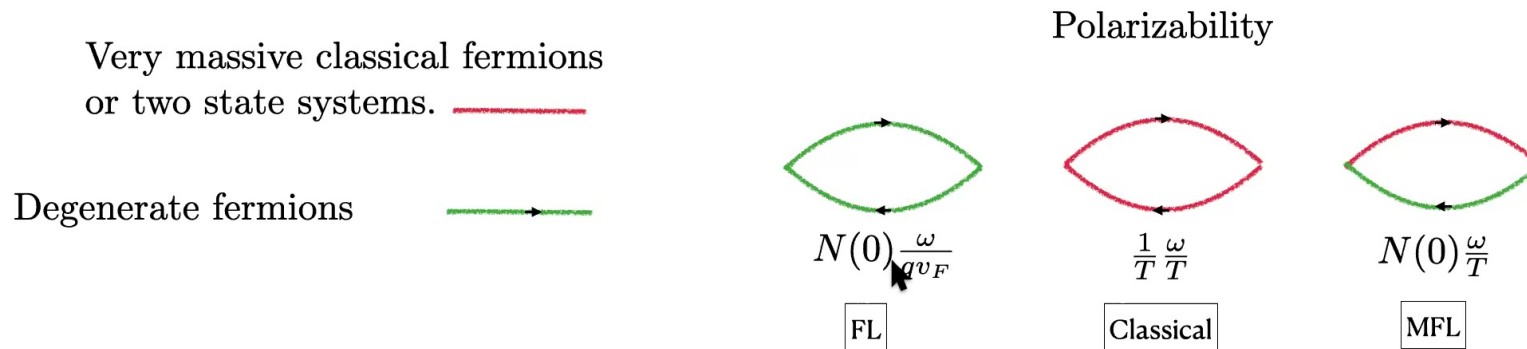
Some but not all the results are similar to numerical results on the Toric model and Kitaev model on the honeycombe lattice for extreme choice of parameters, (seen approximately in Monte-carlo calcs. of Nasu, Udagawa, Motone (PRB-2016) on Kitaev model.) Also Z_2 gauge theory for some unknown range of parameters, etc.





Underlying dispersion theory reason

The numerical calculations are a realization of CMV (1991):
In the energy and temperature range in which some fluctuations are classical and others are nearly degenerate fermions, the absorptive part of the polarizability or mag. susceptibility is $\sim \tanh(\omega/2T)$, essentially independent of momentum.





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Things not understood- (the known un-knowns)



Gives right number for unavailable or dark entropy.
Its insensitivity to substantial magnetic field is probably related to the topological nature of the available states.
Presumably at some ultra low energy scale, R-K kinetic energy could come in so that it is not found at T to 0.

The deviations from perfect Heisenberg interactions on a perfect triangular lattice for the phenomena observed is unknown.

Tuning parameters is required for the log singularity in C/T .
Properties changes as a function of $(m-h)$ smoothly. Low T
 $C/T = \text{constant}$ - crossover from critical Spin-liq.
to (critical) Fermi-liquid ?

Role of Impurities.



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