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UC Davis, Davis

Randomness-induced quantum spin liquid behavior in the s=1/2 Heisenberg antiferromagnet on the pyrochlore lattice

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Quantum spin liquids now observed in many 2D frustrated magnets

RVB state [P.W. Anderson ('73)]

* Triangular lattice *S*=1/2 organic salts

κ-$(ET)_2Cu_2(CN)_3$
$EtMe_3Sb[Pd(dmit)_2]_2$
κ-H_3(Cat-EDT-TTF)$_2$

[K. Kanoda, R. Kato, H. Mori, *et al*]

* Kagome lattice

herbersmithite: $ZnCu_3(OH)_6Cl_2$

[D.G. Nocera *et al*]

* Honeycomb-lattice and square-lattice magnets

6HB- $Ba_3NiSb_2O_9$ (*s*=1)

[J. Quilliam *et al.*, 2016]

$Sr_2CuTe_{1\text{-}x}W_xO_6$

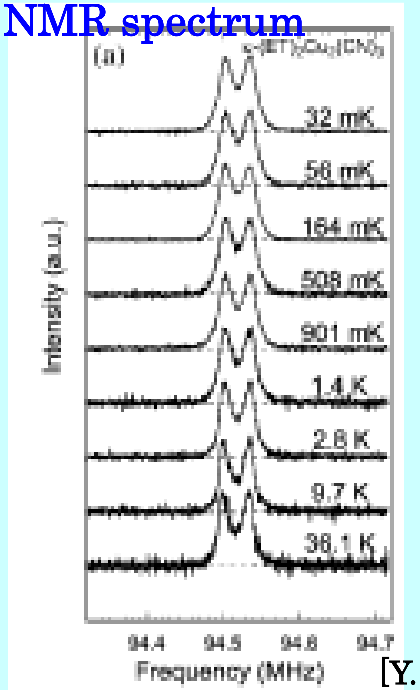
[O. Mustonen *et al.*, 2018]

Competing interactions

Gapless QSL widely observed experimentally

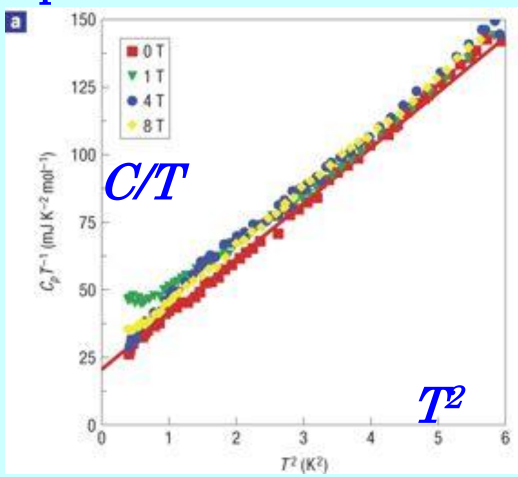
Triangular organic salt κ-ET

NMR spectrum



[Y. Shimizu et al, 2003]

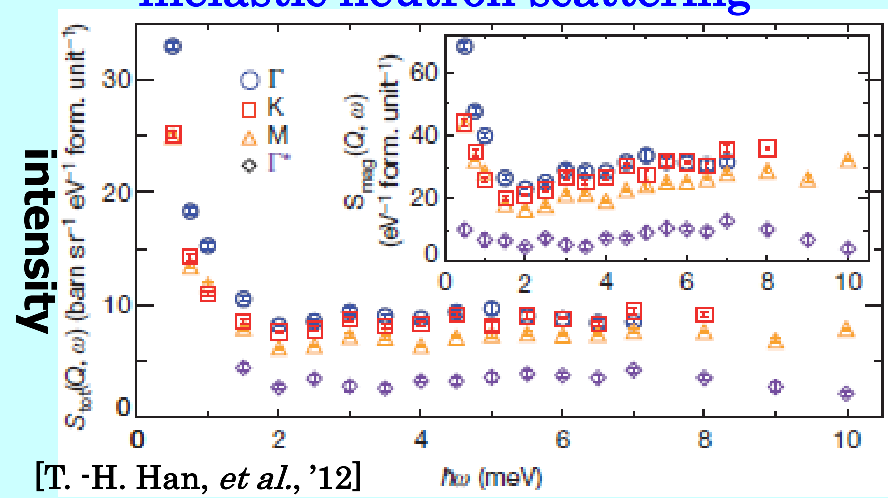
Specific heat



[S.Yamashita, 2008]

Kagome herbertsmithite $ZnCu_3(OH)_6Cl_2$

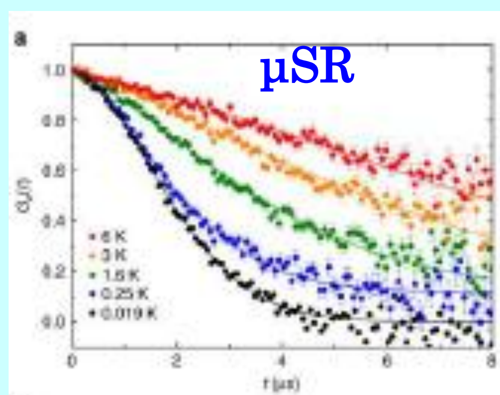
inelastic neutron scattering

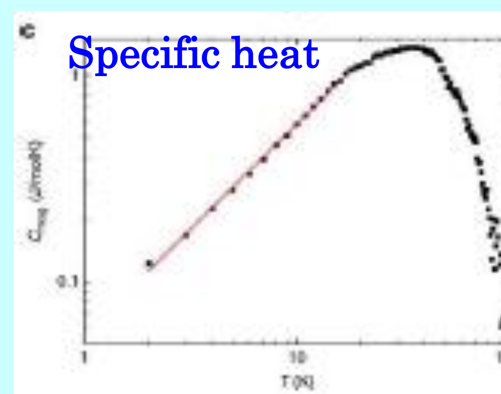


[T. -H. Han, *et al.*, '12]

Square mixed-crystal AF $Sr_2CuTe_{1-x}W_xO_6$

[O .Musutone et al, 2018]



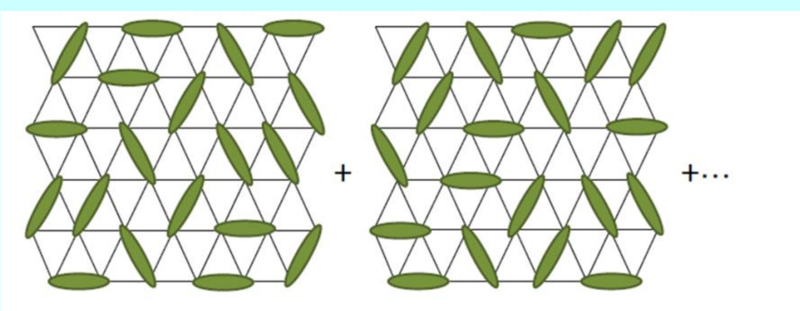


Some (many ?) of experimentally QSL might be randomness-induced ones ?

Randomness (inhomogeneity)

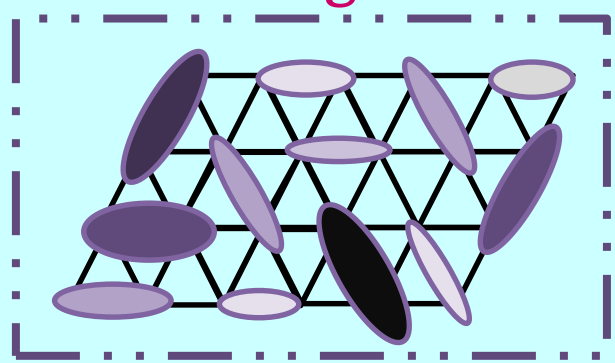
[K. Watanabe, H. Kawamura, H. Nakano and T. Sakai, JPSJ 83, 034714 (2014);
H. Kawamura, K. Watanabe and T. Shimokawa, JPSJ 83, 103704 (2014);
T. Shimokawa, K. Watanabe and H. Kawamura, PRB 92, 134407 (2015)]

“RVB state”



randomness

“random singlet state”



“Anderson localization” of spin singlets ?

Origin of randomness or inhomogeneity could be either extrinsic or intrinsic

Extrinsic randomness

Quenched disorder

* intersite disorder: kagome herbersmithite
* mixed crystal: $Sr_2CuTe_{1\text{-}x}W_xO_6$
* defects, impurities …

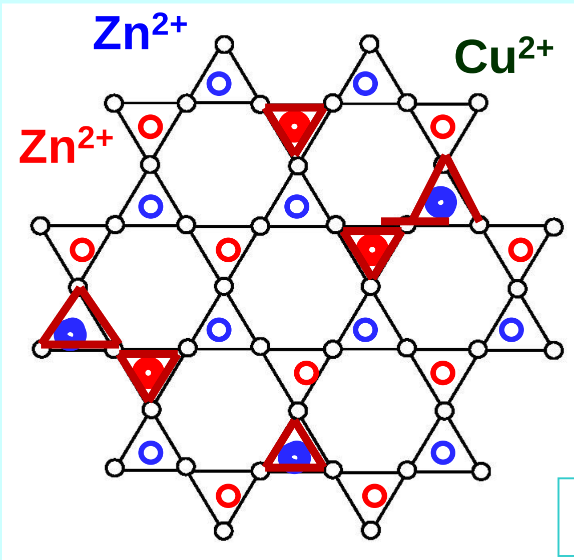
Intrinsic randomness

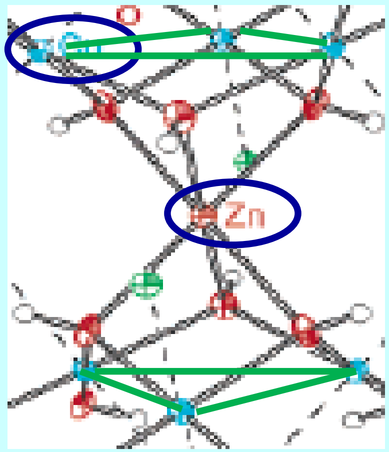
Effective randomness for spin degrees of freedom can be self-generated even in clean systems via the coupling to other degrees of freedom in magnets, e.g., charge, lattices, *etc.*

* coupling to charge (dielectric) degrees of freedom : κ-ET & dmit salts
* coupling to protons at the hydrogen bond: Cat salt

Extrinsic randomness (quenched randomness)

Kagome herbertsmithite: $ZnCu_3(OH)_6Cl_2$





Local Jahn-Teller distortion

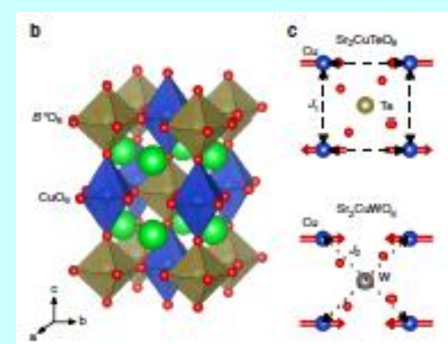
intersite disorder

10~15% Zn^{2+} on the triangular layer randomly replaced by Cu^{2+}

bond-random modulation of the effective exchange J on the kagome plane

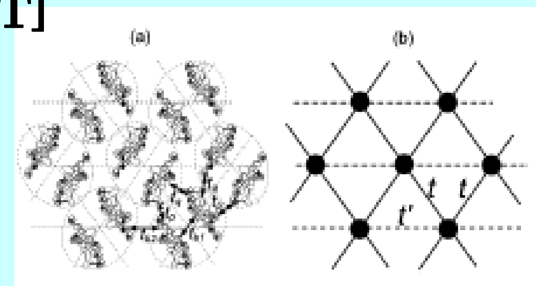
Mixed crystal of square-lattice AFMs: $Sr_2CuTe_{1-x}W_xO_6$

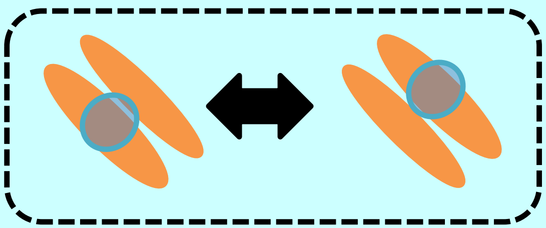
Random s =1/2 J_1-J_2 square-lattice Heisenberg AF
Sr_2CuTeO_6 (J_2/J_1 = 0.03, T_N = 29K)
and Sr_2CuWO_6 (J_2/J_1 = 7.92, T_N = 24K)
⇒ Significant quenched disorder associated with the Te/W occupation

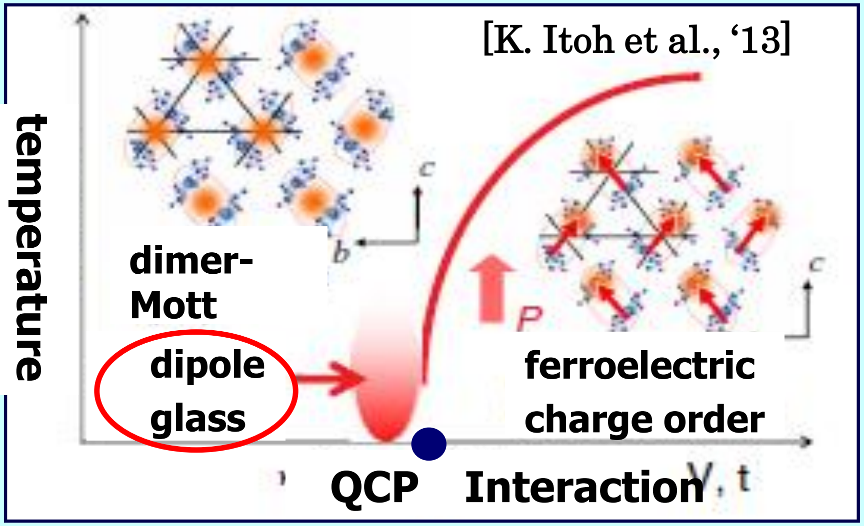


Relevant randomness (inhomogeneity) exists in trianglar organic salts ?

[κ-ET]

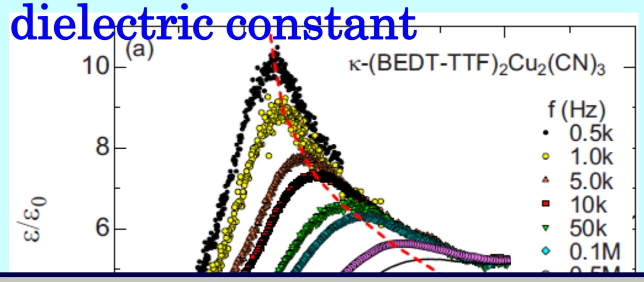




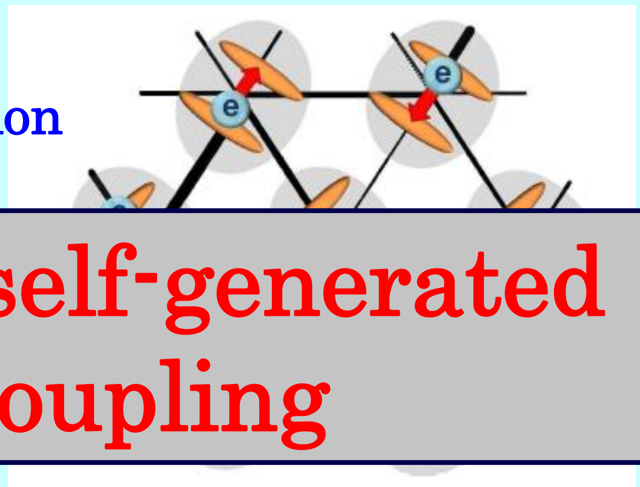


Charge inhomogeneity
dynamically self-generated

AC dielectric constant



spatial
modulation
of *Jij*



Effective randomness is self-generated via the spin-charge coupling

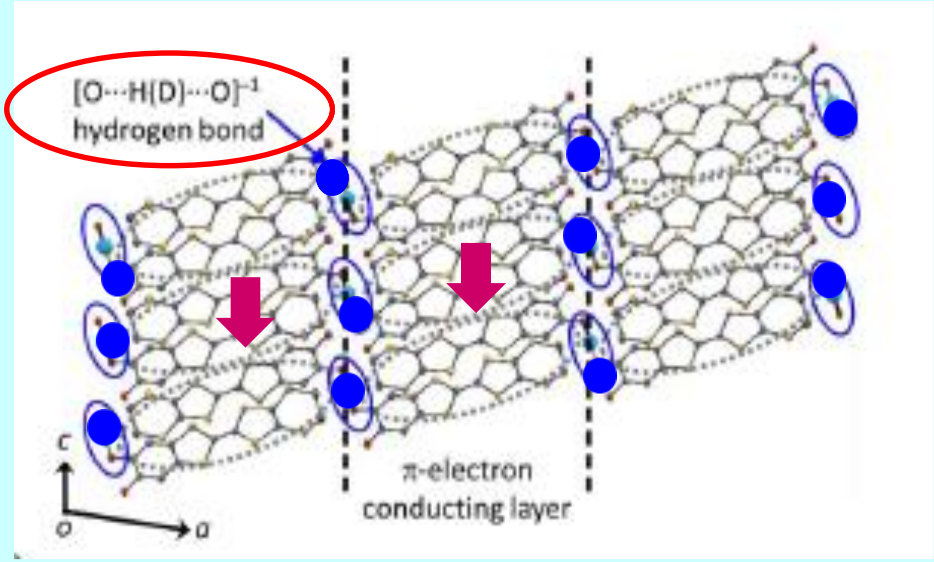
The “third” quantum spin liquid

κ-H_3(Cat-EDT-TTF)$_2$

[H. Mori’s group, 2013]

π-electron - proton coupled triangular organic conductor

hydrogen-bonded



Proton remains delocalized

[T. Tsumuraya et al, 2015]

→ possibly slowed down into random positions at low-T

→ yielding random fields to π–electrons

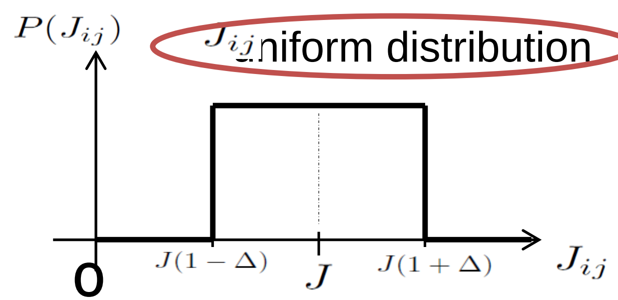
→ spatially modified random exchange coupling J_{ij}

➡ Gapless random-singlet state

Bond-random S=1/2 AF Heisenberg model on the triangular & kagome lattices

$$\mathcal{H} = \sum_{<i,j>} J_{ij} \hat{S}_i \cdot \hat{S}_j$$

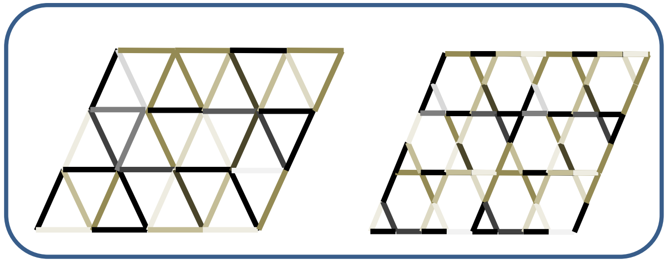
$$\left(\ 0 \leq J(1-\Delta) \leq J_{ij} \leq J(1+\Delta) \ \right)$$



Δ : randomness parameter （0 ≦ Δ ≦ 1 ）

$\Delta = 0$ → **no randomness**

$\Delta = 1$ → **maximal randomness**



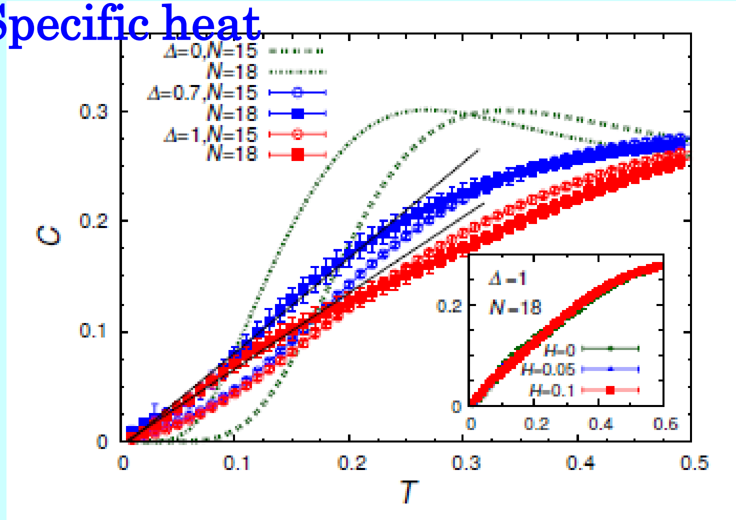
Exact diagonalization (ED) calculation performed on various 2D models, including triangular, kagome, J_1-J_2 honeycomb and square lattices

➡ find a QSL-like state (random-singlet state)

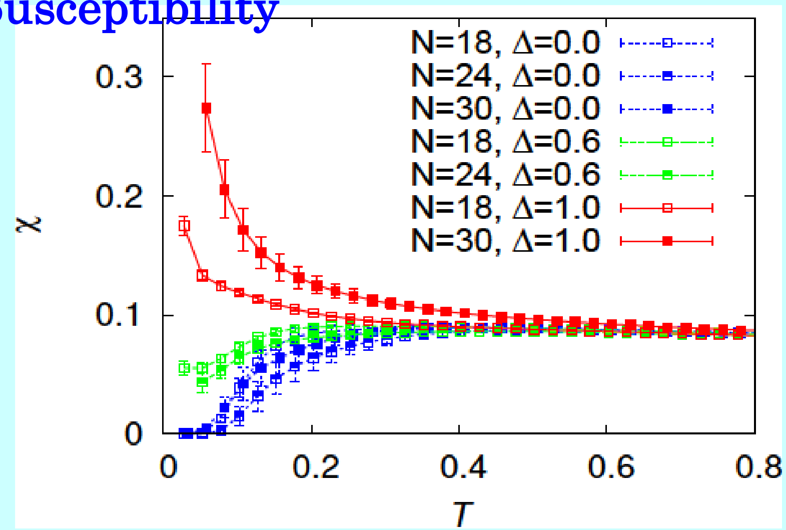
ED numerical results on 2D models

Random triangular model

Specific heat

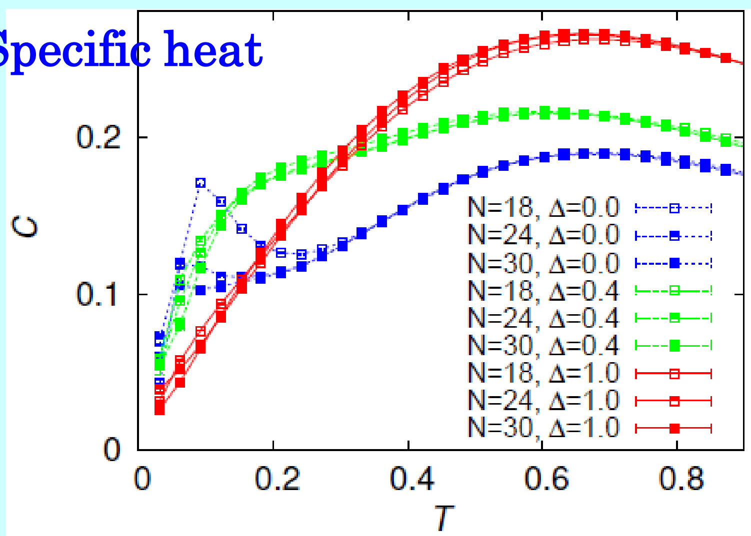


Susceptibility

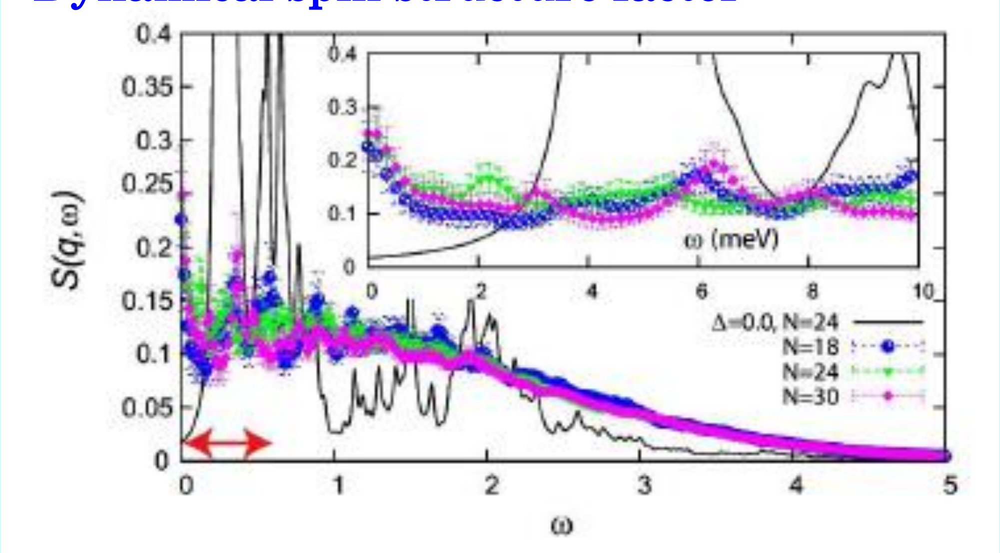


Random kagome model

Specific heat



Dynamical spin structure factor



Randomness-induced QSL state
--- random-singlet state ---
appear to be realized in
a variety of 2D frustrated magnets

Randomness-induced QSL state
ever possible in 3D ?
***e.g.*,** pyrochlore ?

Gapless QSL behavior observed in pyrochlore AF: $Lu_2Mo_2O_5N_2$

[L. Clark, … A. Harrison, J. P. Attfield, B.D. Gaulin, PRL 113, 117201 (2014)]

$Lu_2Mo_2O_7$

Mo^{4+} $4d^2$ $S=1$ → Orbital degrees of freedom

Apparently disorder-free system

Spin-glass order at $T_f=16$K (similarly to $Y_2Mo_2O_7$)

due to the spin-orbital coupling

Spin glass

$Lu_2Mo_2O_5N_2$

Random subsitution of O^{2-} by N^{3-}

Mo^{5+} $4d^1$ $S=1/2$

$S=1/2$ pyrochlore Heisenberg AF

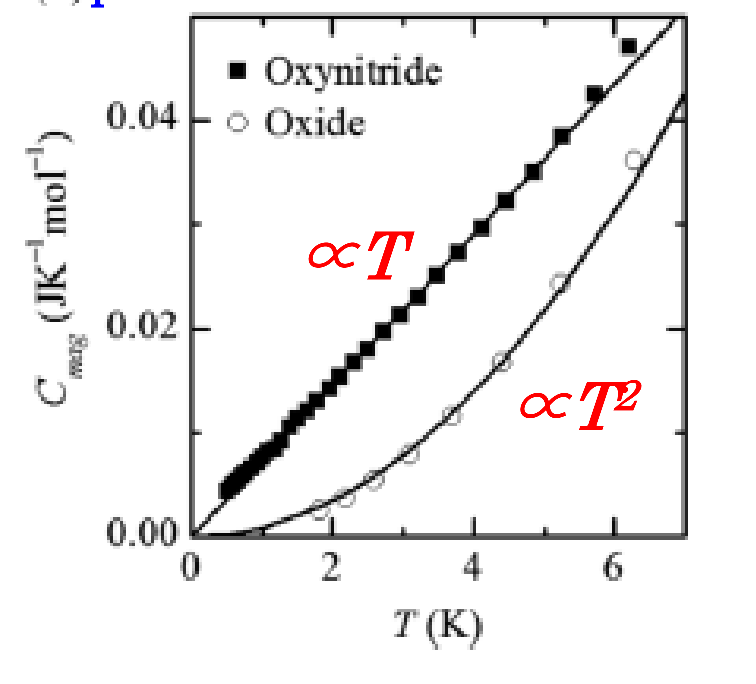
with significant exchange randomness

QSL !

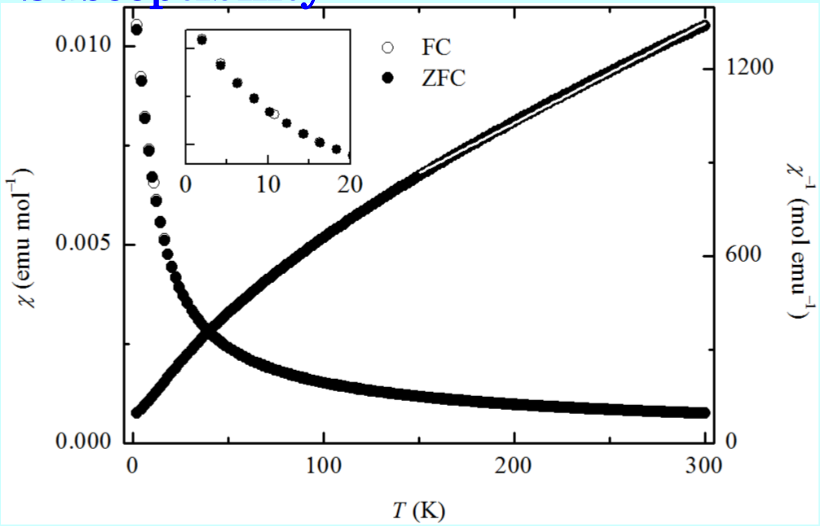
QSL behavior of $Lu_2Mo_2O_5N_2$

[L. Clark *et al.*, 2014]

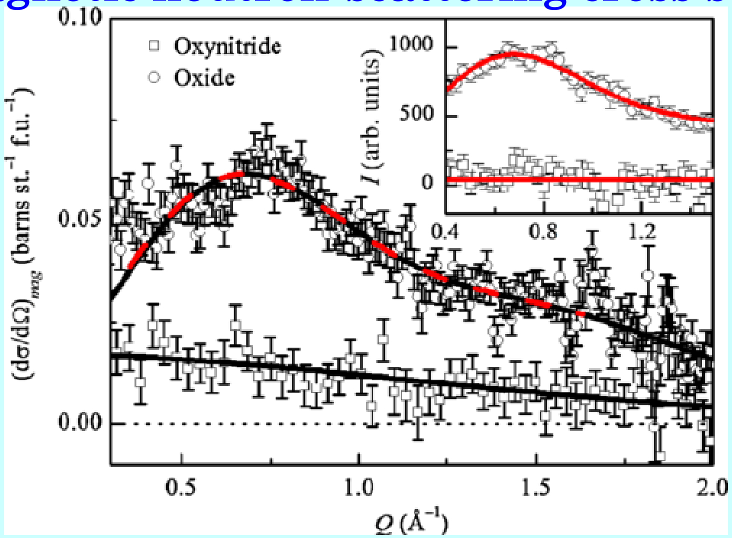
Specific heat



Susceptibility

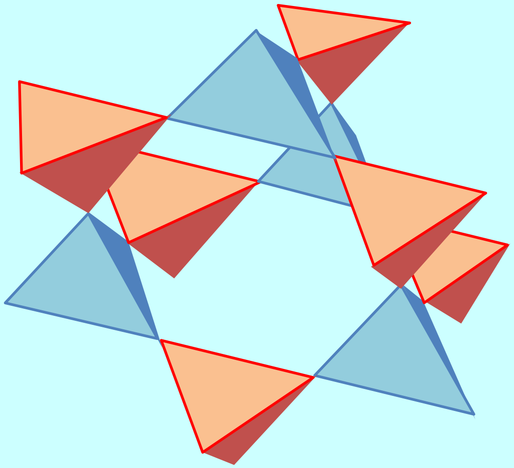


Magnetic neutron-scattering cross section

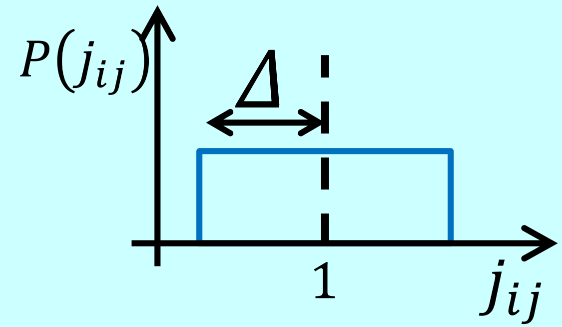


Gapless QSL behavior with the T–linear specific heat and broad features in the spin structure factor

Model: $s = 1/2$ random-bond AF Heisenberg model on the 3D pyrochlore lattice



$$\mathcal{H} = \sum_{\langle i,j \rangle} J_{ij} \, \vec{S_i} \cdot \vec{S_j}$$



* Nearest-neighbor coupling
* Periodic boundary conditions

* ED (ground state properties, $N \leq 36$)
& Hams-de Raedt method (finite-temperature properties, $N = 32$)

* Averaged over 10~100 samples

---

* Preceding works on the regular model

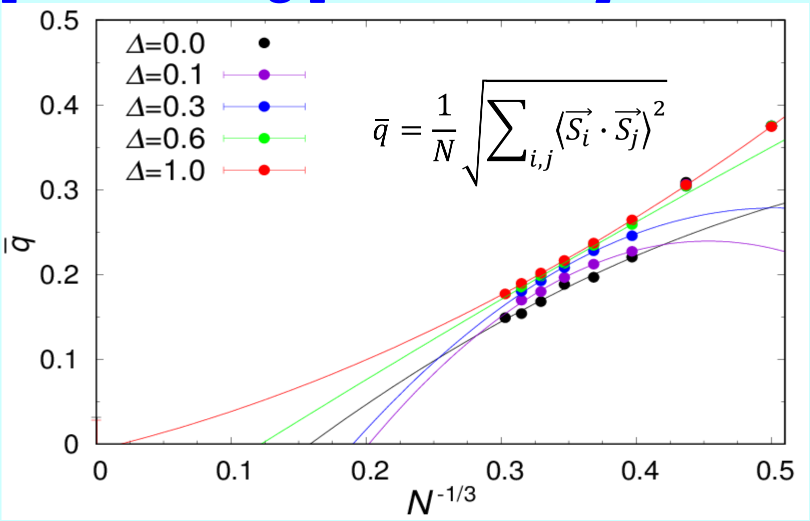
some sort of non-magnetic state [1-7]

- Valence Bond Crystal ? [1-4]
- Chiral Spin Liquid ? [5,6]
- something else ? [7]

[1] A. B. Harris *et al.*, J. Appl. Phys. 69, 5200 (1991).
[2] M. Isoda and S. Mori, J. Phys. Soc. Jpn. 67, 4022 (1998).
[3] H. Tsunetsugu, J. Phys. Soc. Jpn. 70, 640 (2001).
[4] E. Berg *et al.*, Phys. Rev. Lett. 90, 147204 (2003).
[5] J. H. Kim and J. H. Han, Phys. Rev. B 78, 180410 (2008).
[6] F. J. Burnell *et al.*, Phys. Rev. B 79, 144432 (2009).
[7] V. R. Chandra and J. Sahoo, Phys. Rev. B 97, 144407 (2018).

Results: Ground state properties

Spin freezing parameter q



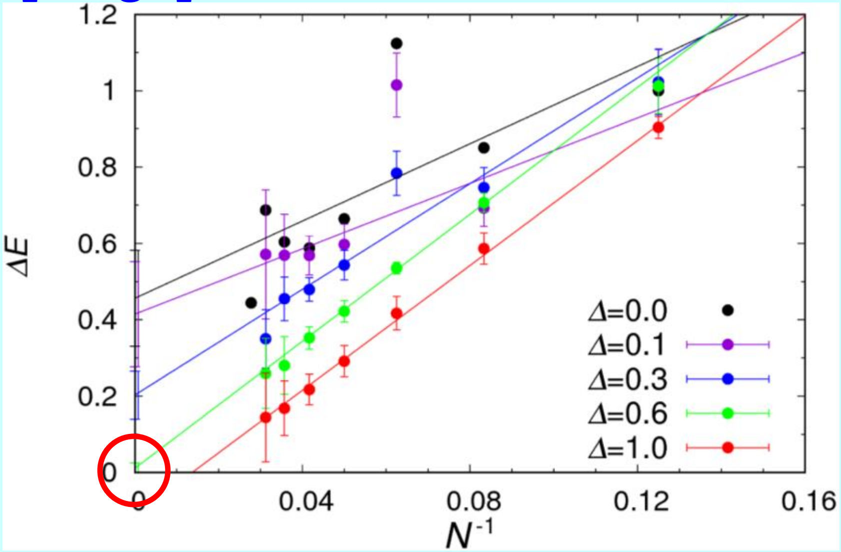
No magnetic order at any Δ

Seems to be gapful for $\Delta < \Delta_c \sim 0.6$
but gapless for $\Delta > \Delta_c$

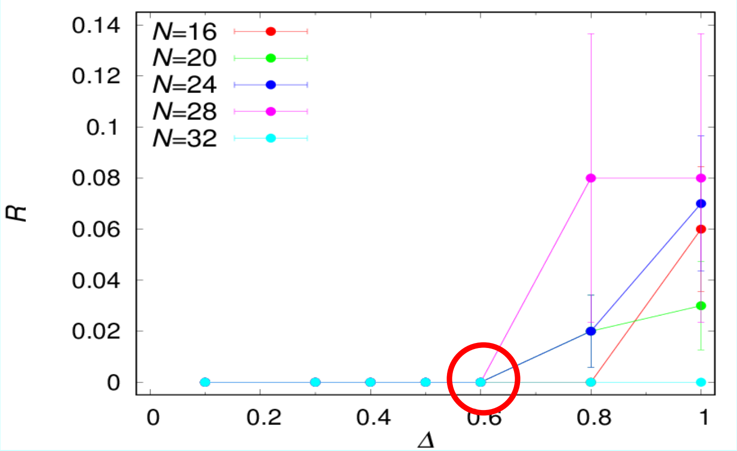
Gapful-gapless transition
at $\Delta = \Delta_c \sim 0.6$

Random singlet state realized
at $\Delta = \Delta_c \sim 0.6$

Spin gap

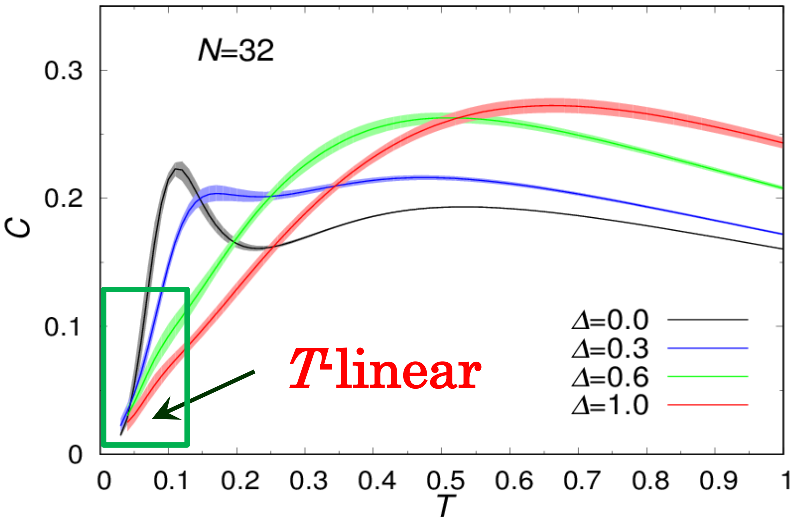


Rate of triplet ground states

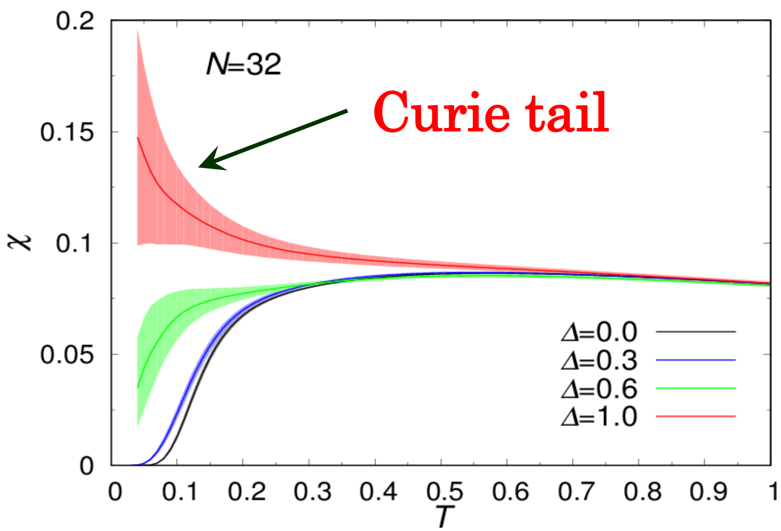


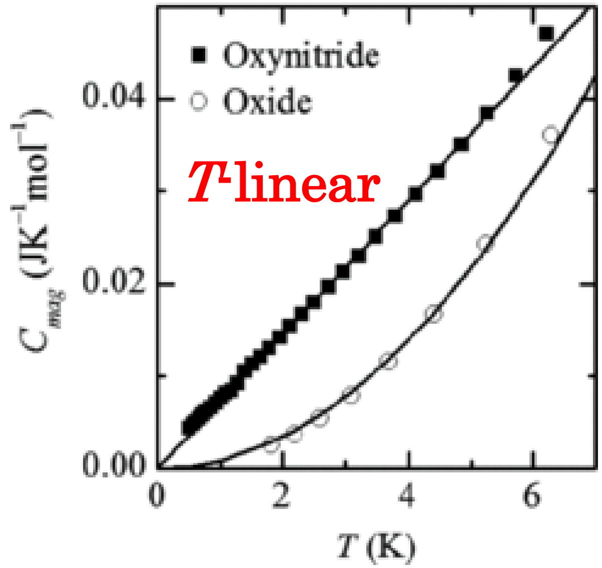
Finite-temperature properties

Specific heat



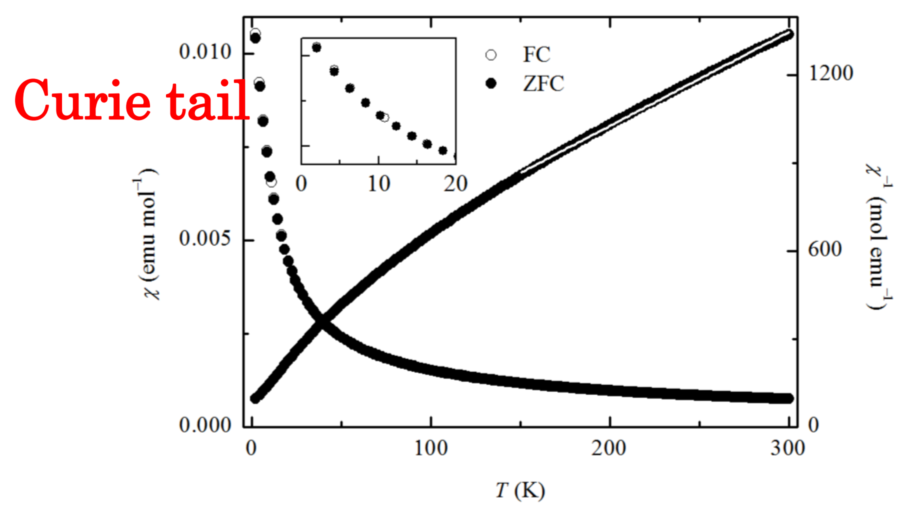
Susceptibility





$Lu_2Mo_2O_5N_2$ (exp.)

[L. Clark *et al.*, 2014]

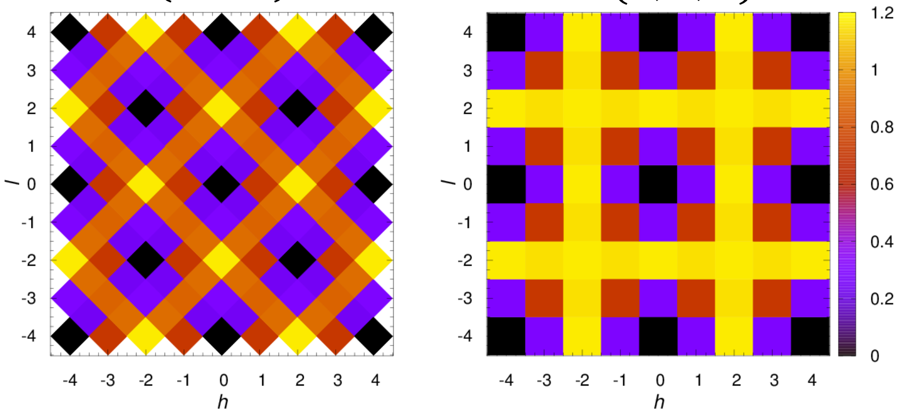


Static spin correlations (T=0)

Static spin structure factor *S(q)*

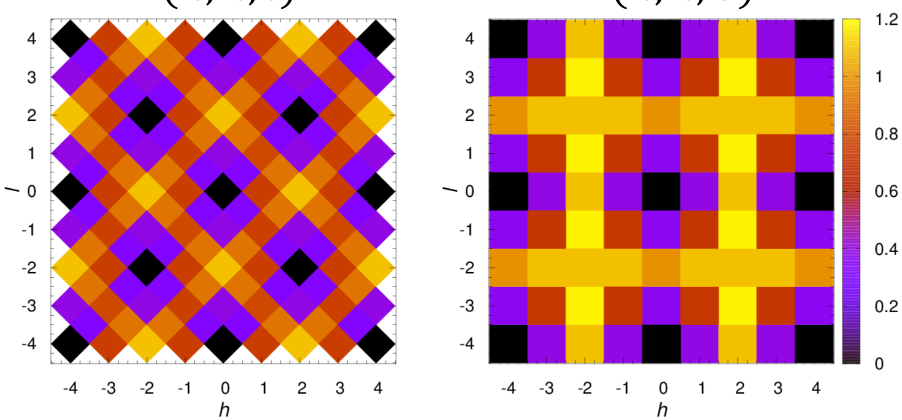
Regular: $\Delta = 0$

(h, h, l) $(h, k, 0)$

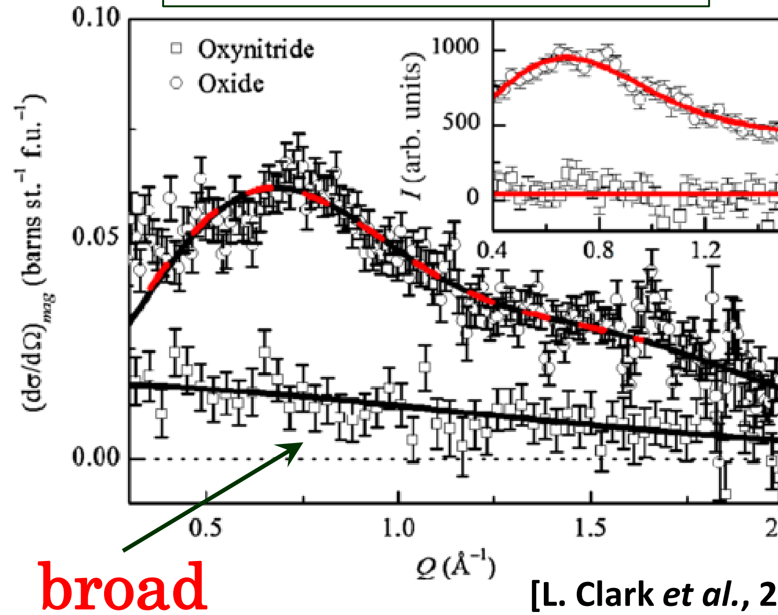


Random: $\Delta = 1$

(h, h, l) $(h, k, 0)$



$Lu_2Mo_2O_5N_2$ (exp.)



[L. Clark *et al.*, 2014]

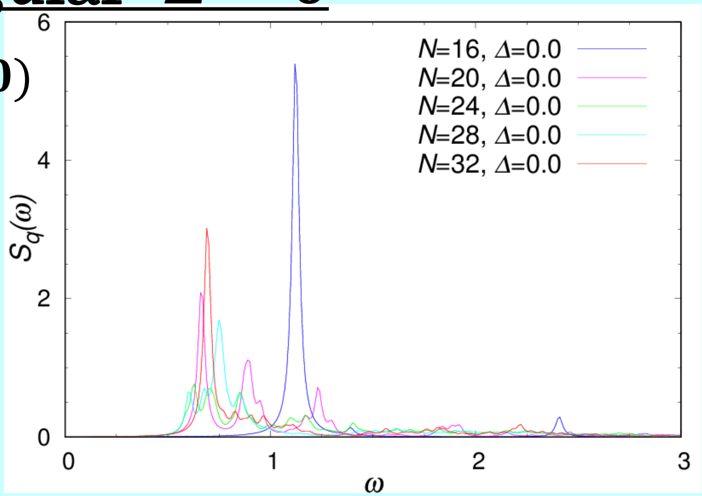
Very broad spin structure factors without any peaky structure

Dynamical spin correlations (T=0)

Dynamical spin structure factor $S(q,\omega)$

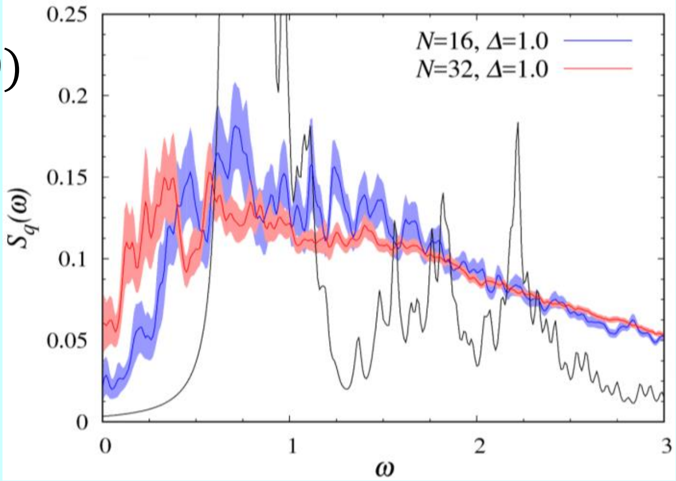
Regular: $\Delta = 0$

$(2, 2, 0)$

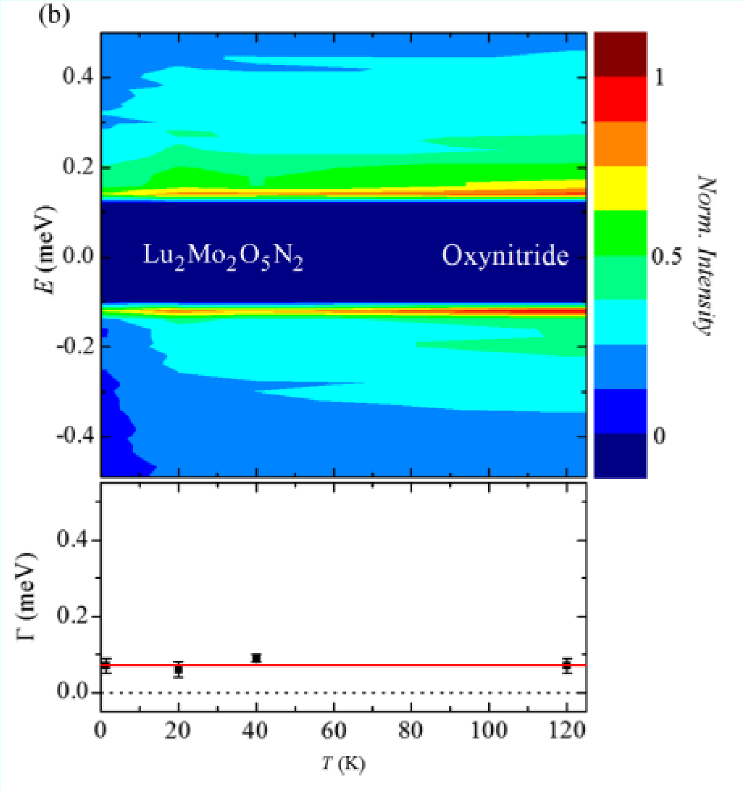


Random: $\Delta = 1$

$(2, 2, 0)$



$Lu_2Mo_2O_5N_2$ (exp.)

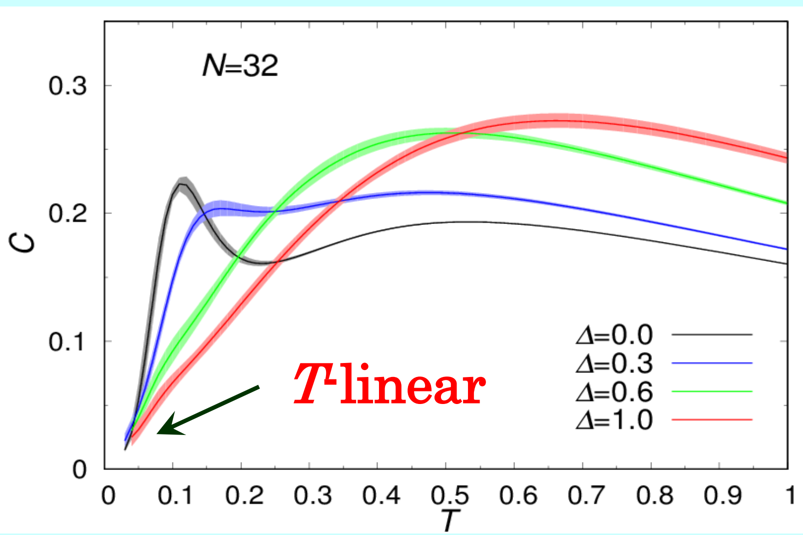


[L. Clark *et al.*, 2014]

Random-singlet state in 3D looks similar to the one in 2D

e.g., specific heat

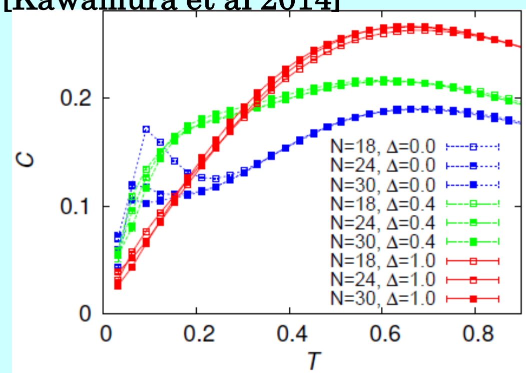
3D pyrochlore



2D models

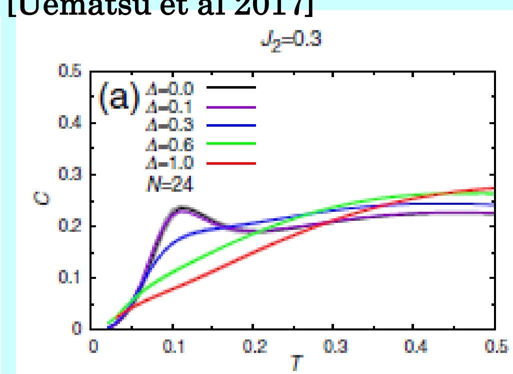
kagome

[Kawamura et al 2014]



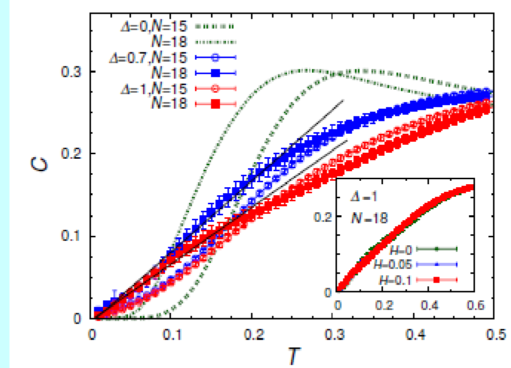
J_1-J_2 honeycomb

[Uematsu et al 2017]



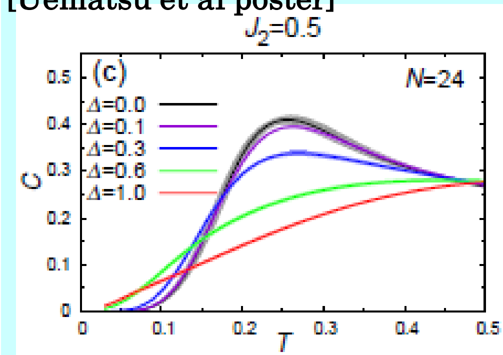
triangular

[Watanabe et al 2014]



J_1-J_2 square

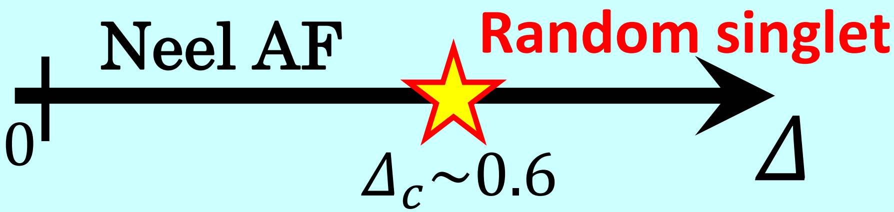
[Uematsu et al poster]



3D Pyrochlore

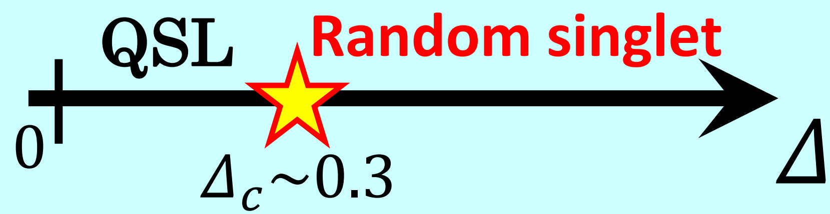
◆ triangular

[Watanabe *et al*, 2014]



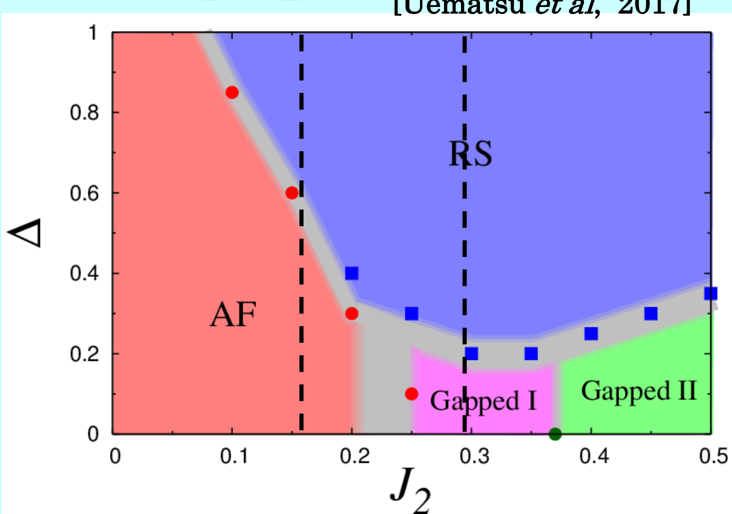
◆ kagome

[Kawamura *et al*, 2014]



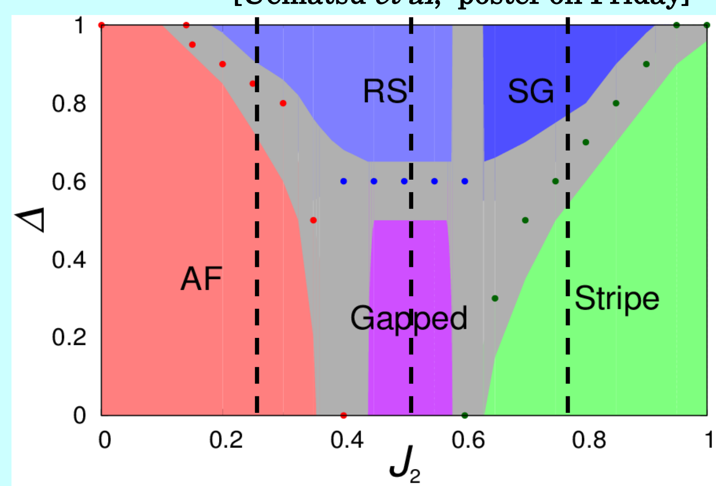
◆ J_1-J_2 honeycomb

[Uematsu *et al*, 2017]

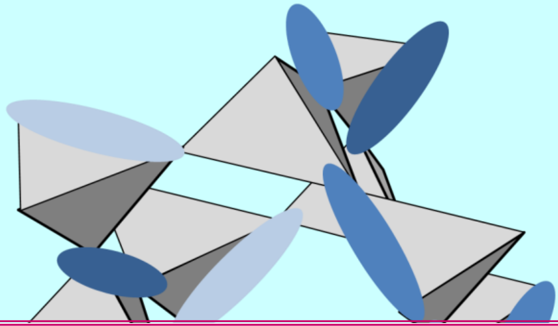


◆ J_1-J_2 square

[Uematsu *et al*, poster on Friday]



Summary

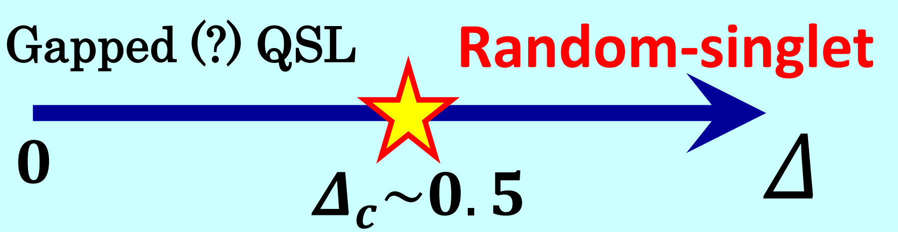


* Zero-T and finite-T properties of the bond-randm $s=1/2$ AF Heisenberg model

Randomness or inhomogeneity plays a role in quantum magnetism !

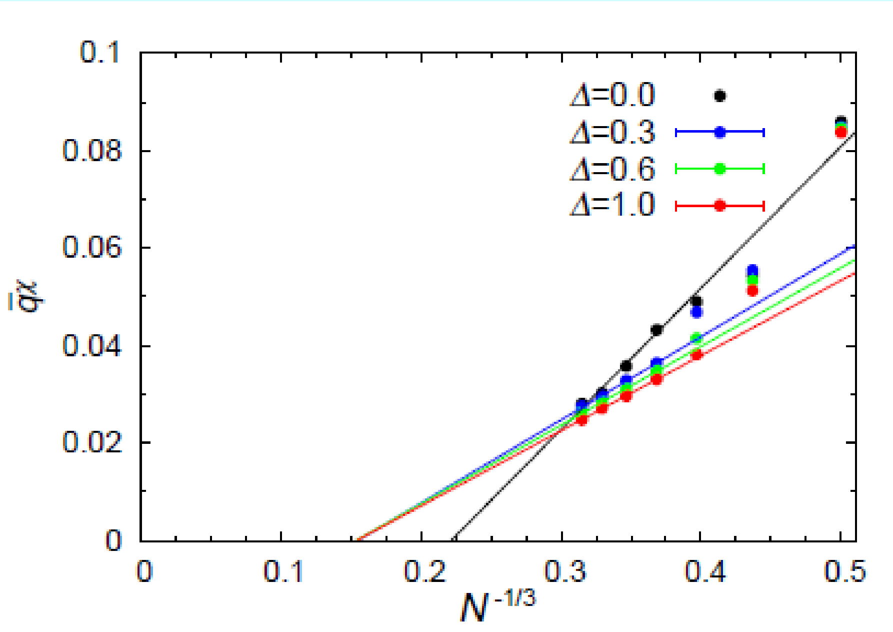
* The random-singlet state in 3D is very much similar to the one in 2D, characterized by the T−linear low-T specific heat, gapless susceptibility with a Curie-like tail, and broad features in the spin structure factor.

* The results are consistent with the recent experimental result on the pyrochlore AF $Lu_2Mo_2O_5N_2$

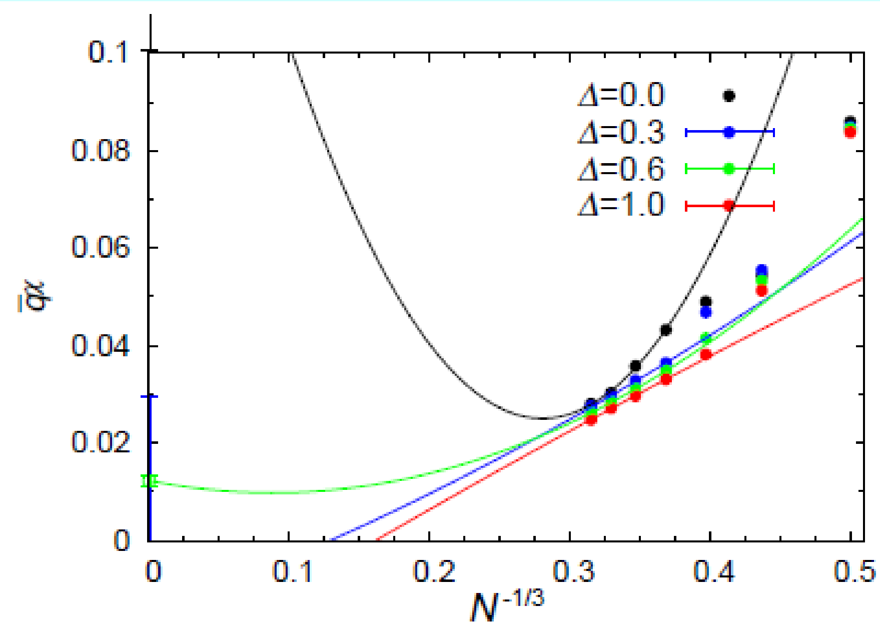


Possible chiral order ?

Scalar chirality

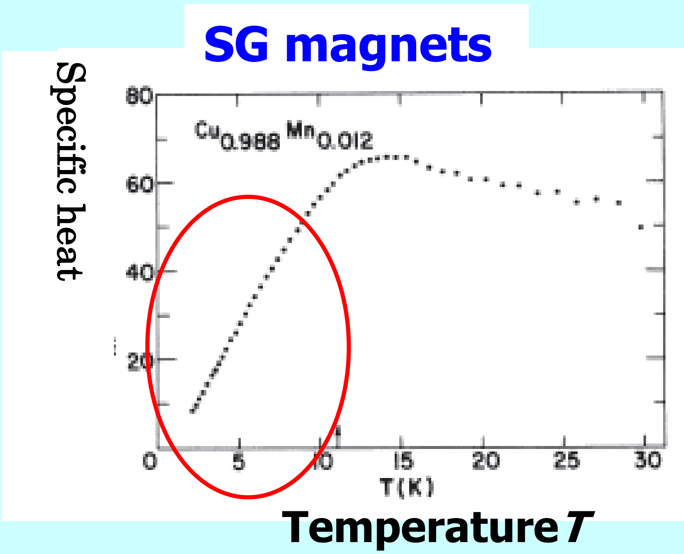


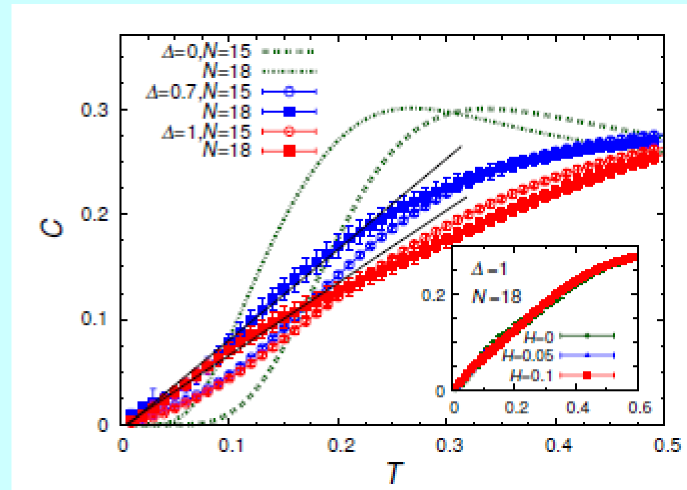
Vector chirality



No chiral(-glass) order
both in the regular and the random cases

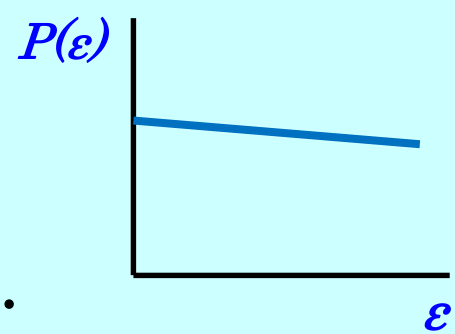
Origin of the T–linear specific heat in the random-singlet state





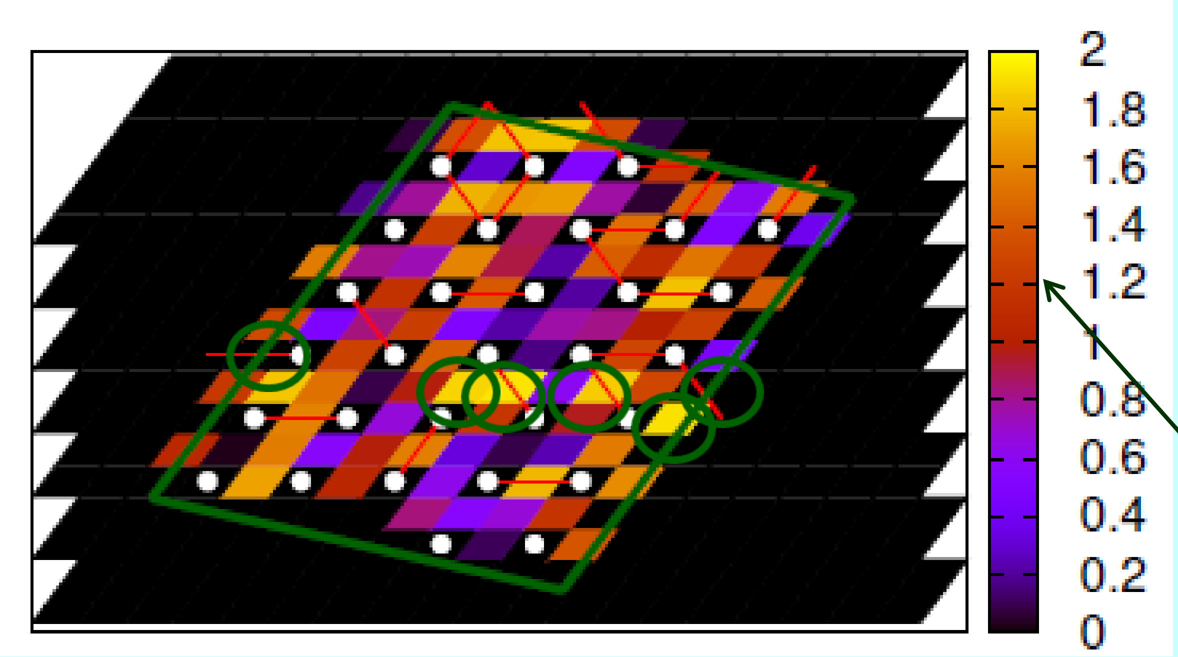
T-linear specific heat generically realized in spin glasses and molecular glasses, reflecting continuous low-energy excitations with a nonzero density of states down to zero.

[P.W. Anderson et al, '72]



Nature of the "random-singlet" state

Anderson-localized RVB state ?



bond with a strong singlet large $-\langle Si \cdot Sj \rangle$

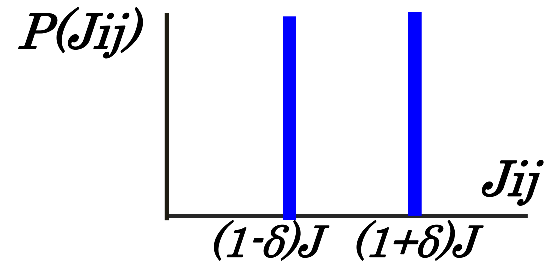
J_{ij} strength

A subtle balance between the kinetic energy (resonance) and the potential energy (random J_{ij})

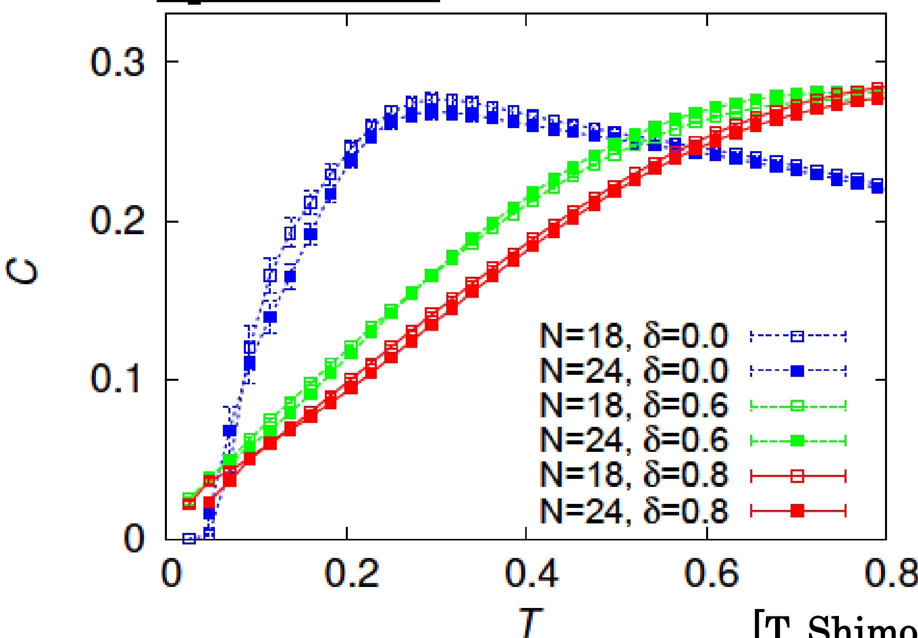
Gapless behavior robust against $P(J_{ij})$?

Yes.

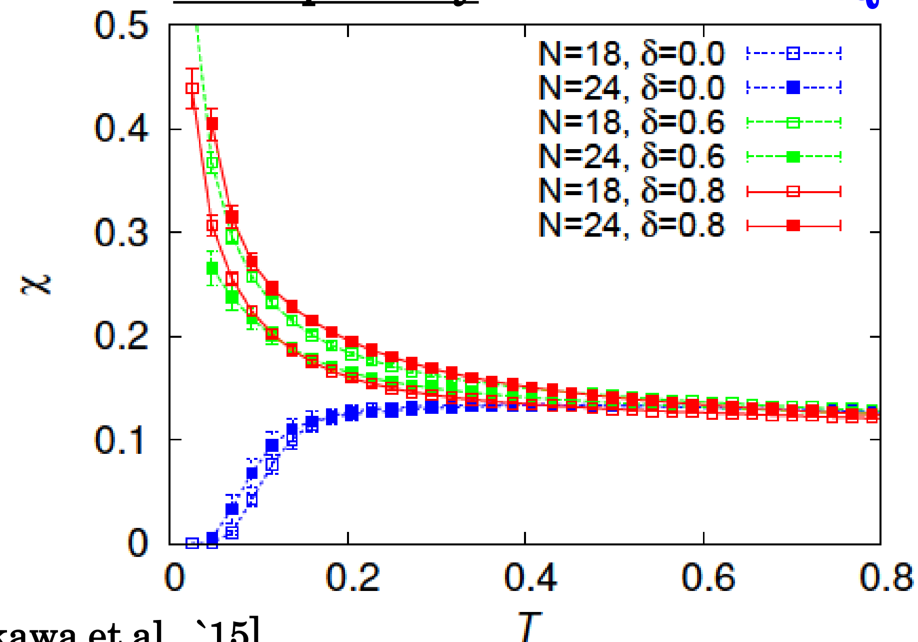
[Ex.] Discrete (binary) J_{ij} distribution



Specific heat TPQ



Susceptibility TPQ



[T. Shimokawa et al., `15]

Gapless behavior for larger randomness (δ) even for the discrete (binary) J_{ij} distributin