

Destruction of magnetic long-range order by quenched disorder: Triangular & Pyrochlore AFM

Matthias Vojta
(TU Dresden)

Santanu Dey
Eric Andrade
Jose Hoyos
Stephan Rachel

(Dresden)
(Sao Paulo)
(Sao Paulo)
(Melbourne)

Destruction of magnetic long-range order by quenched disorder: Triangular & Pyrochlore AFM

1. Frustration + quenched disorder \rightarrow spin glass?

2. **Triangular-lattice Heisenberg antiferromagnet**

Dipolar spin texture from bond defect

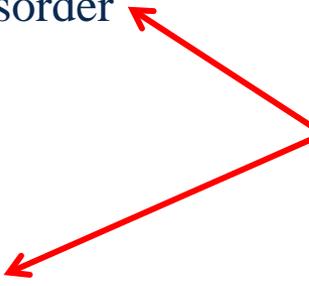
Destruction of long-range order for infinitesimal disorder

3. **Pyrochlore-lattice XY antiferromagnet**

Order by disorder

Destruction of long-range order for finite disorder

**by defect-induced
transverse fields**



Frustration & disorder

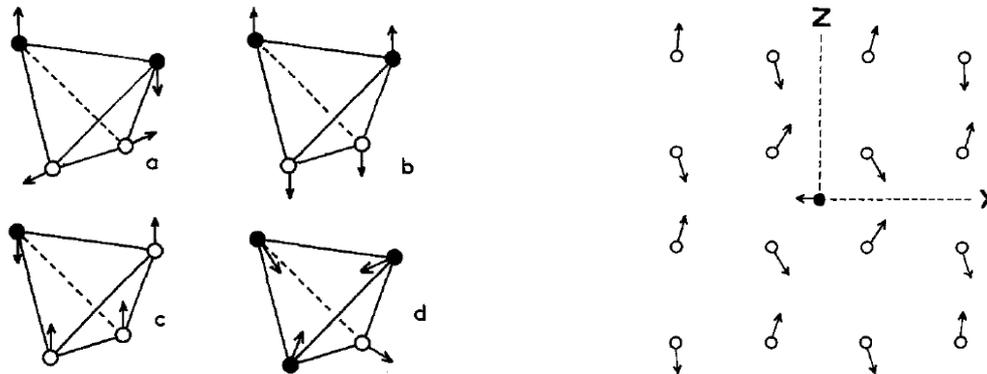
Insulating Spin Glasses

Jacques Villain

Département de Recherche Fondamentale, Laboratoire de Diffraction Neutronique,
Centre d'Etudes Nucléaires de Grenoble, Grenoble, France

Received September 21, 1978

The possibility of obtaining spin glasses by addition of impurities in an antiferromagnetic insulator is examined. Dipolar interactions are briefly considered but the attention is focussed on Heisenberg systems. Equivalence with the Edwards-Anderson model is derived in a theoretical case. Experimental realisations, such as quasi-one dimensional systems, and spinels, are reviewed. A weak concentration of non-magnetic impurities can give rise to a new state that we call "semi spin glass", in which a ferromagnetic component coexists with a transverse, spin glass component. An important case is when the pure system has a high ground state degeneracy (cooperative paramagnet). Non-magnetic impurities or other forms of disorder can transform it into a spin glass.



Quenched disorder

Bond randomness, random vacancies, ... \rightarrow Fate of ordered state?

Order of clean system
stable

Randomness induces
different ordered state

Long-range order destroyed
in favor of ...

spin glass

random-singlet
state

Weak randomness - two cases:

- 1) Gapped phase – stable against weak randomness (defects screened)
- 2) Gapless phase – less clear

Triangular lattice Heisenberg AFM

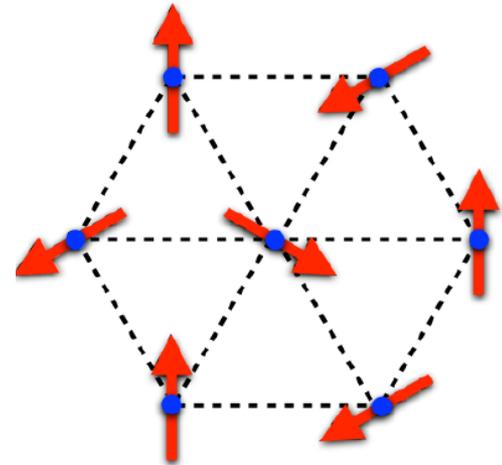
+

quenched disorder

Triangular Heisenberg antiferromagnet

$$H = J_1 \sum_{\langle ij \rangle} \vec{S}_i \cdot \vec{S}_j + J_2 \sum_{\langle\langle ij \rangle\rangle} \vec{S}_i \cdot \vec{S}_j$$

Couplings $\alpha = J_2/J_1 < 1/8$



Spin stiffness in classical limit:

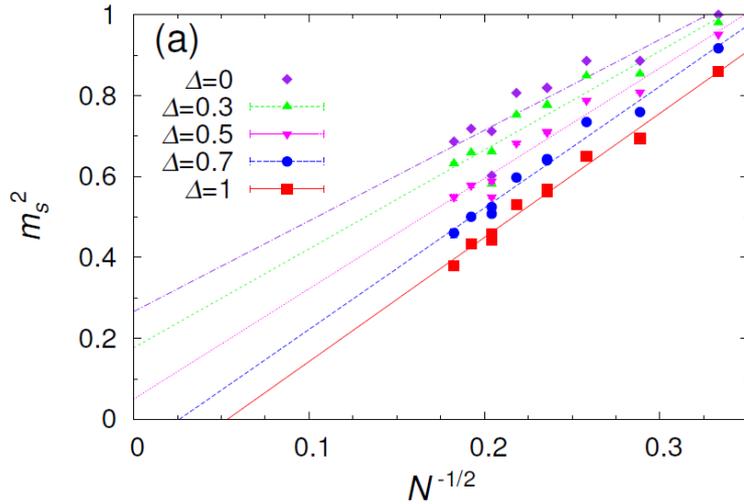
$$\rho_s^{\parallel} = N_0^2 (J_1 - 6J_2) \frac{\sqrt{3}}{2} \mathcal{A}$$

↑
order parameter amplitude $N_0=1$

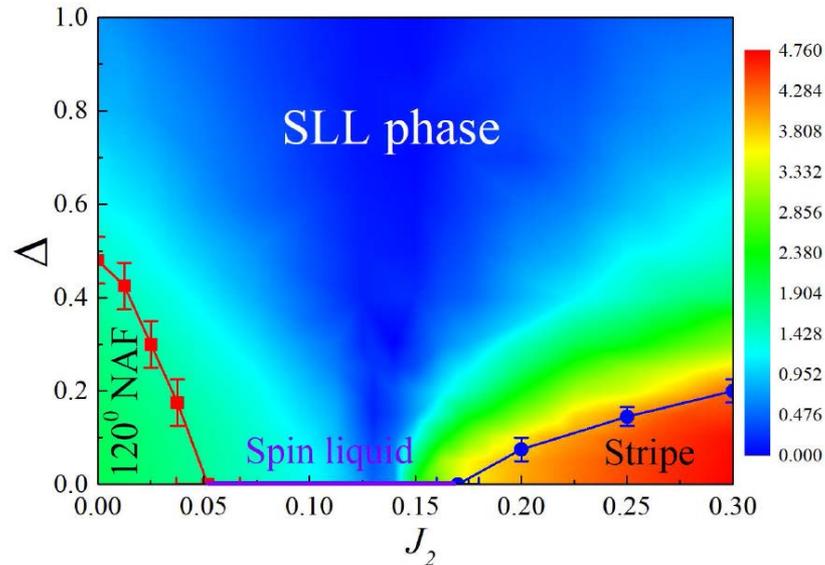
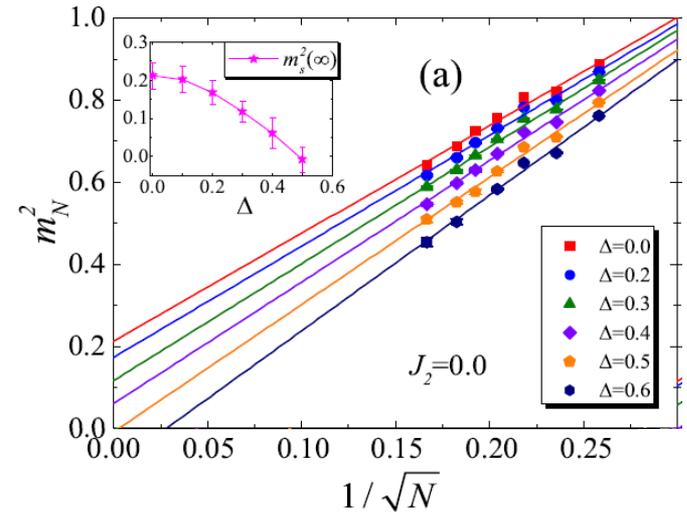
↙
area of unit cell

Triangular Heisenberg antiferromagnet + bond disorder

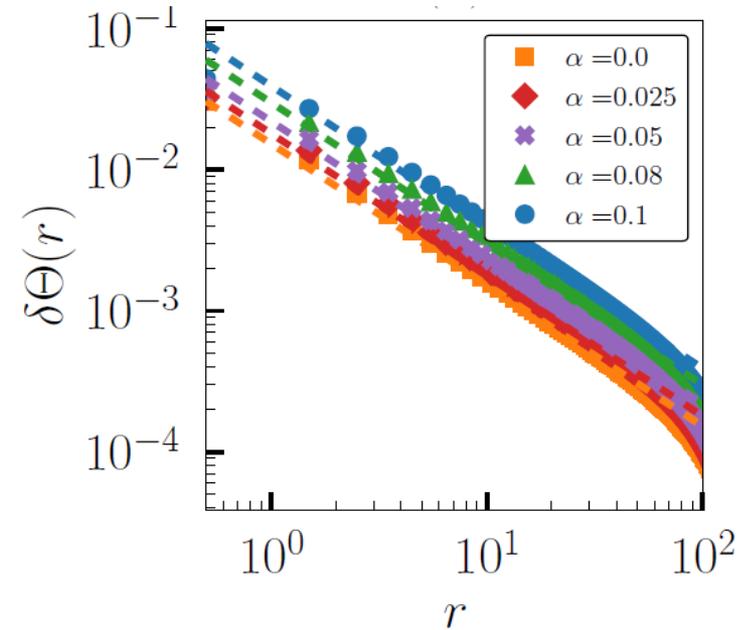
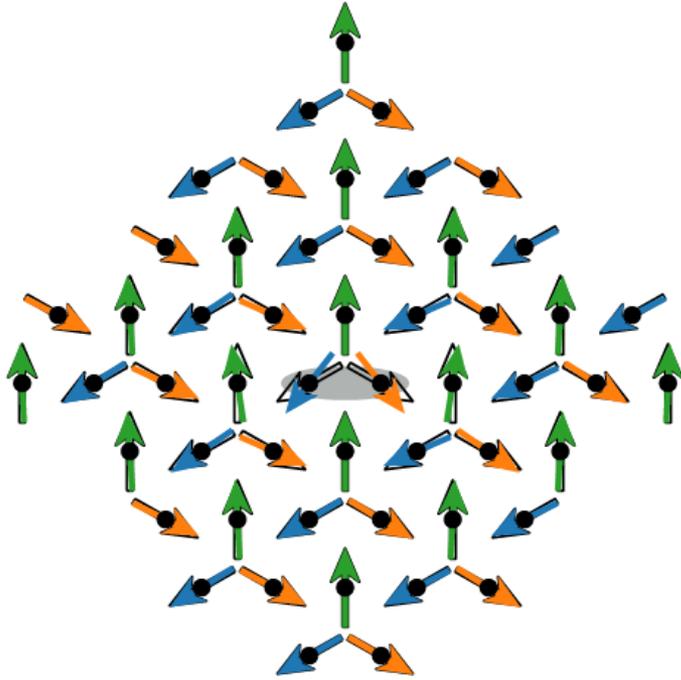
$S=1/2$, exact diagonalization,
box disorder of strength Δ



$S=1/2$, ED + DMRG,
box disorder of strength Δ



Bond defect in triangular Heisenberg AFM



Linear-response theory for defect-induced texture:

$$\langle S_i^\perp \rangle = \lambda(\delta J) N_0 \mathcal{A} \int \frac{d^d q}{(2\pi)^d} (i\hat{e} \cdot \vec{q}) \chi^\parallel(\vec{q}) e^{i\vec{q} \cdot \vec{r}_i}$$

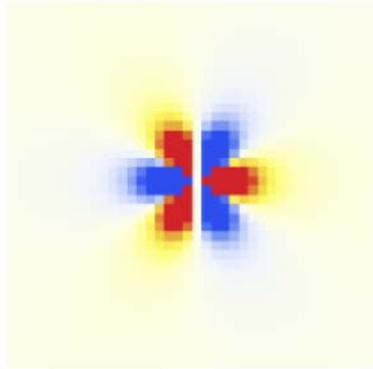
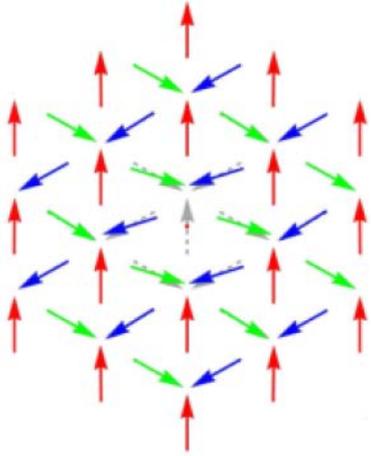
State remains coplanar, with dipolar texture:

$$\delta\Theta(\vec{r}) = \kappa \delta J \frac{N_0^2}{\tilde{\rho}_s} \frac{\hat{e} \cdot \vec{r}}{r^d}$$

N_0 : order parameter
 $\tilde{\rho}_s$: clean-limit stiffness
 \hat{e} : defect bond vector

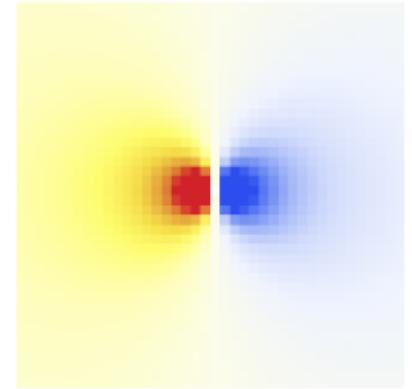
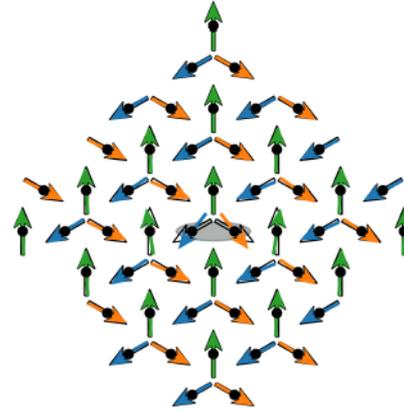
Site vs. bond disorder in triangular AFM

vacancy \rightarrow octupole



$$\delta\Theta \sim 1/r^3$$

bond defect \rightarrow dipole



$$\delta\Theta \sim 1/r$$

Finite defect concentration: Fate of long-range order

Superposition of textures from random dipoles d_{ij} at locations r_{ij} :

$$\langle S_l^\perp \rangle = \kappa \frac{N_0^3}{\tilde{\rho}_s} \sum_{\langle ij \rangle} \frac{\vec{d}_{ij} \cdot \vec{r}_{l,ij}}{r_{l,ij}^d}$$

Disorder-averaged „transverse“ magnetization (Δ : disorder strength):

$$\overline{\langle S_l^{\perp 2} \rangle} = \tilde{\kappa} \Delta^2 \frac{N_0^6}{\tilde{\rho}_s^2} \int dr r^{1-d}$$

Fluctuations diverge for $d \leq 2$ (!)

**Destruction of non-collinear LRO by infinitesimal bond disorder
for $d \leq 2$**

Destruction of long-range order



Conjecture: Resulting state is **spin glass** with finite correlation length ξ

Estimate ξ as domain size from stability condition $\overline{\langle S_l^\perp{}^2 \rangle} \lesssim N_0^2$

$$\xi^{2-d} \propto \tilde{\rho}_s^2 / (\Delta^2 N_0^4)$$

$d=2$ is **marginal** case, with exponentially large ξ for weak disorder Δ :

$$\ln \frac{\xi}{\xi_\infty} \propto \frac{\tilde{\rho}_s^2}{\Delta^2 N_0^4}$$

RG analysis

Non-linear sigma model for order parameter R (rotation matrix, $\vec{S}_i = R_i \cdot \vec{N}_i$):

$$\mathcal{S} = -\frac{\tilde{\rho}}{4} \int d\tau d^d x \left[\frac{1}{c^2} \text{Tr} (R^{-1} \partial_\tau R)^2 + \text{Tr} P (R^{-1} \partial_i R)^2 \right]$$

Add bond disorder:

$$\delta J_{ij} \vec{S}_i \cdot \vec{S}_j = \delta J_{ij} \vec{N}_i \cdot R^{-1} (\hat{e}_{ij} \cdot \nabla) R \cdot \vec{N}_j$$

Replicas, disorder average, one-loop RG:

$$\text{Stiffness} \quad \beta(\eta_a) = \frac{d\eta_a}{d \log b} = (1-d)\eta_a + \frac{R_{abc}}{8\pi} \left[\left(\frac{\eta_a^2}{\eta_b \eta_c} \right) + \frac{2\eta_a^2 \sigma_c}{\eta_b} \right]$$

$$\text{Disorder} \quad \beta(\sigma_a) = \frac{d\sigma_a}{d \log b} = (d-2)\sigma_a - \frac{R_{abc}}{8\pi} \left[\left(\frac{\sigma_b}{\eta_c} + \frac{2\eta_a \sigma_a}{\eta_b \eta_c} \right) + 2(\sigma_b \sigma_c + \frac{2\eta_a \sigma_a \sigma_c}{\eta_b}) \right]$$

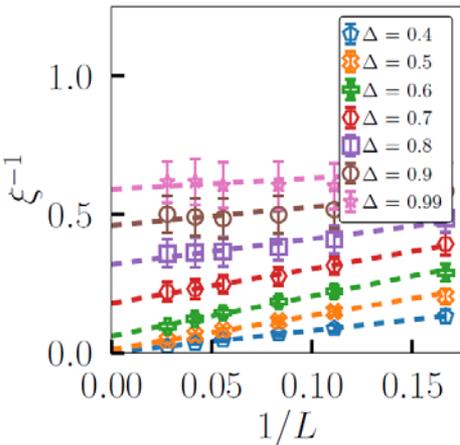
**Destruction of non-collinear LRO by infinitesimal bond disorder
for $d \leq 2$**

Numerical results

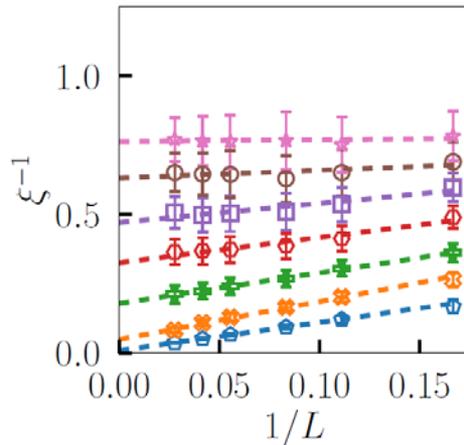
$$\alpha = J_2/J_1$$

Finite-size scaling of correlation length ξ

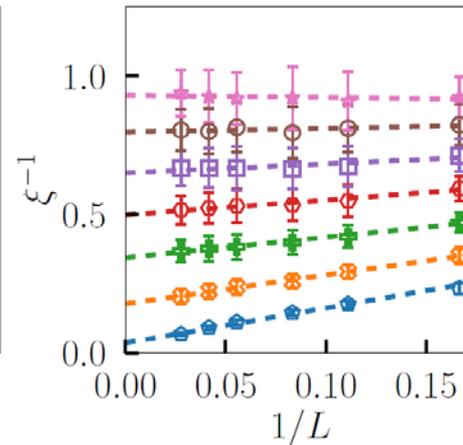
(a) $\alpha = 0.0$



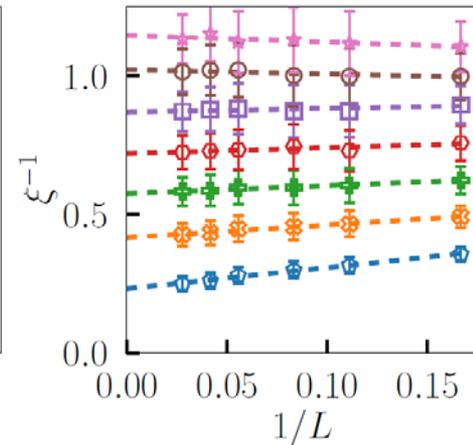
(b) $\alpha = 0.025$



(c) $\alpha = 0.05$

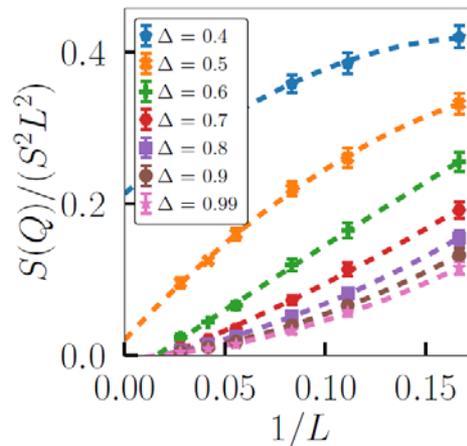


(d) $\alpha = 0.08$

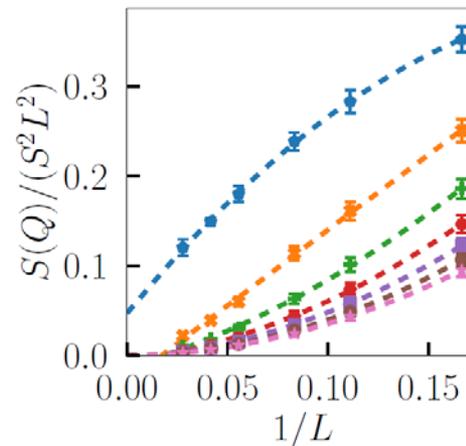


Finite-size scaling of order parameter

(a) $\alpha = 0.025$

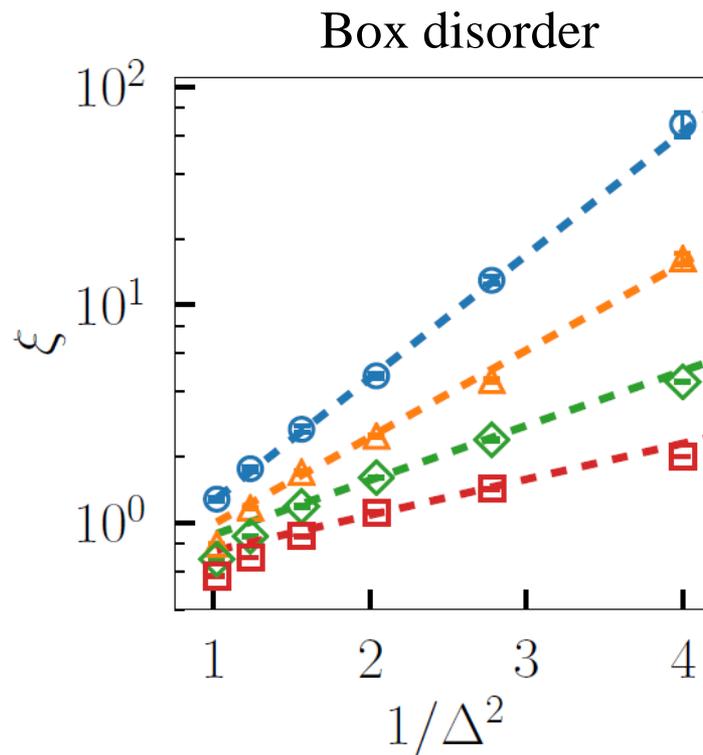
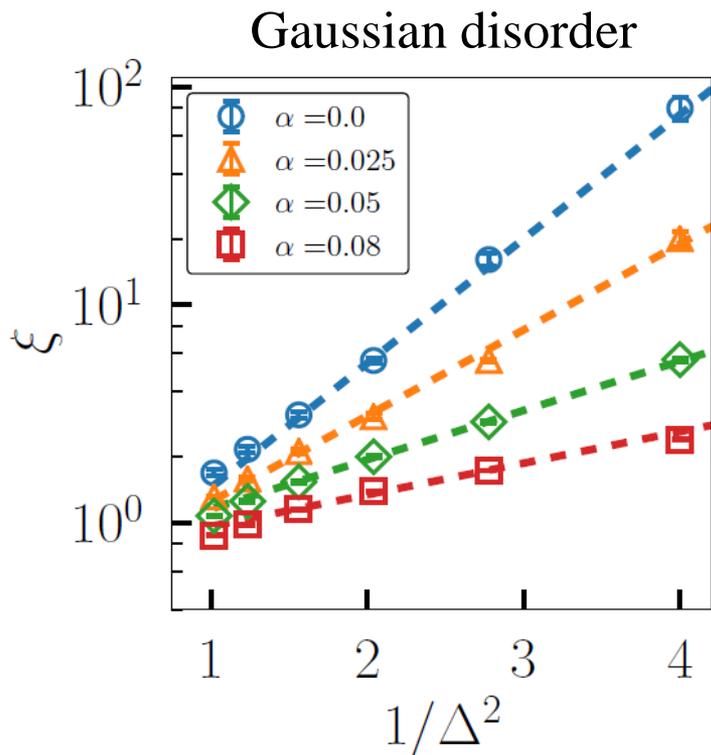


(b) $\alpha = 0.05$

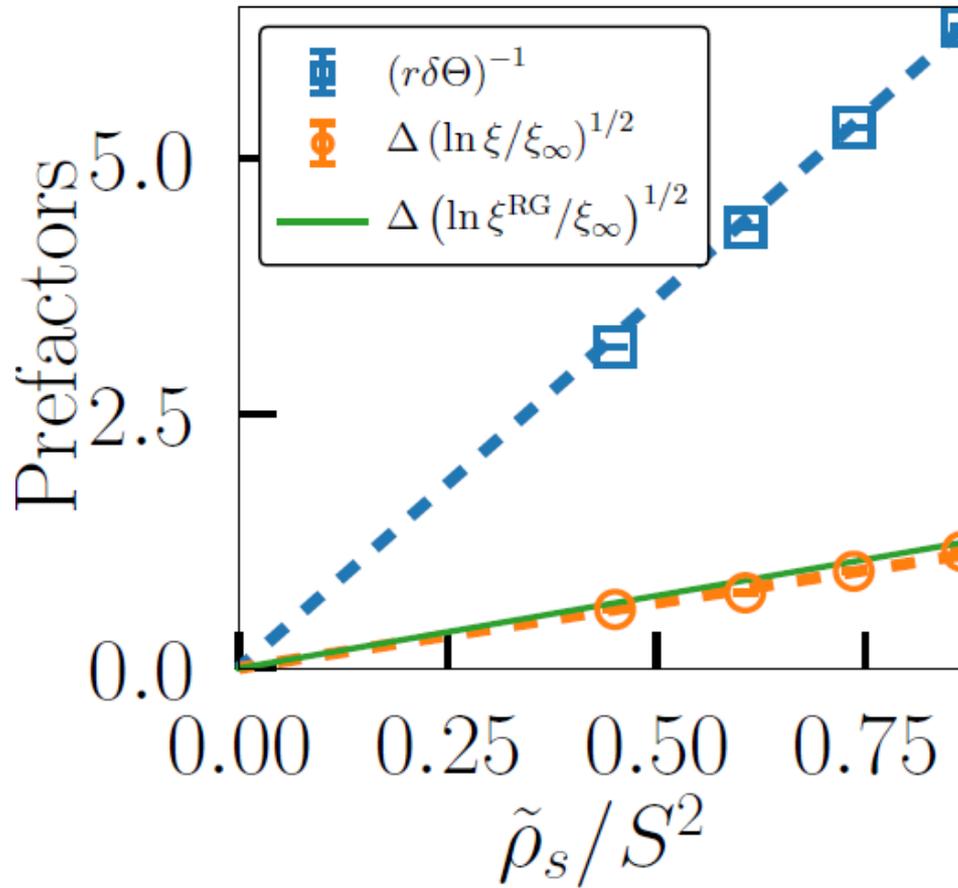


Numerical results: Correlation length

Recall prediction ($d=2$): $\ln \frac{\xi}{\xi_\infty} \propto \frac{\tilde{\rho}_s^2}{\Delta^2 N_0^4}$



Numerical results: Stiffness dependence



Amplitude of dipolar texture

$$\delta\Theta(\vec{r}) = \kappa \delta J \frac{N_0^2}{\tilde{\rho}_s} \frac{\hat{e} \cdot \vec{r}}{r^d}$$

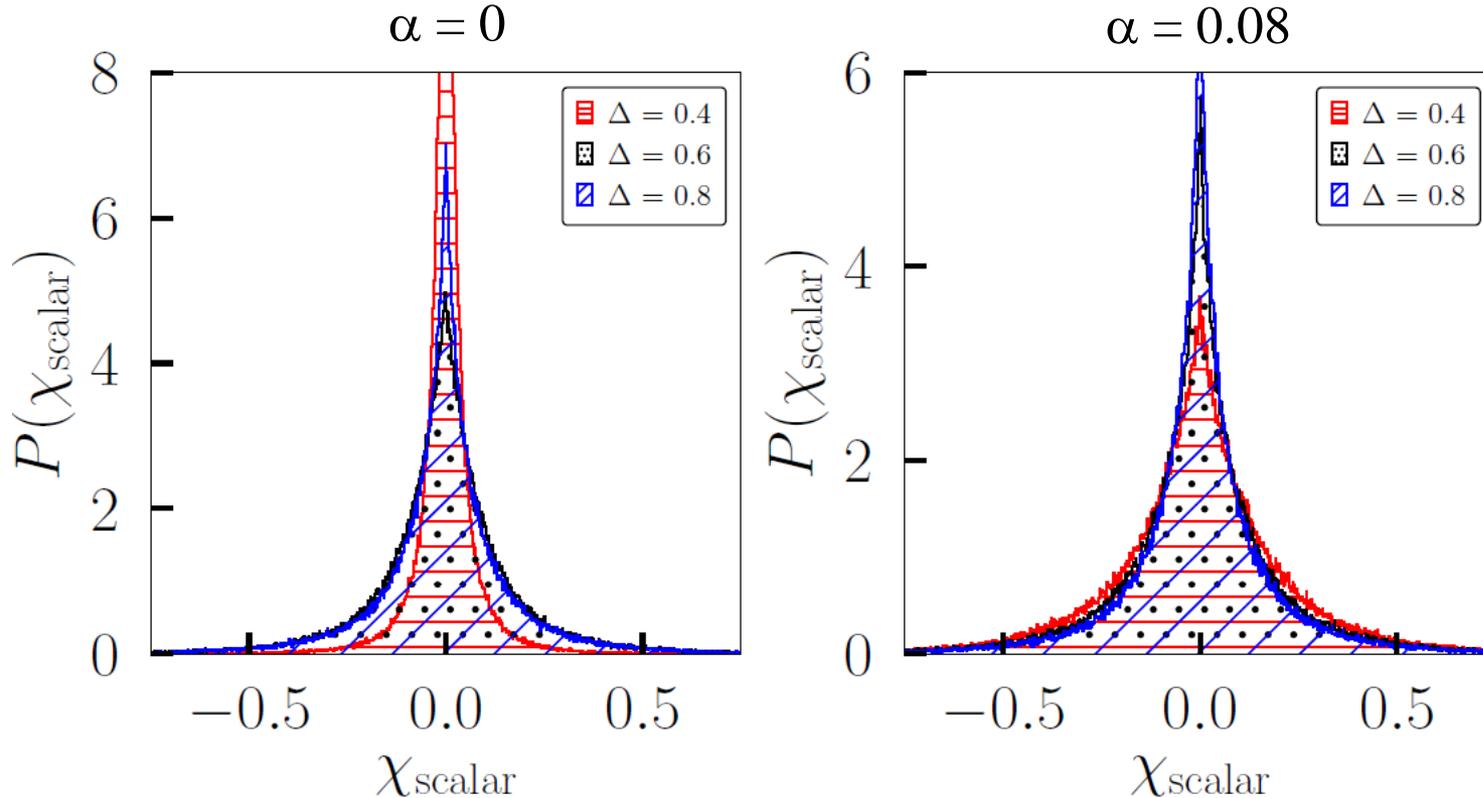
Prefactor in exponential for corr.length

$$\ln \frac{\xi}{\xi_\infty} \propto \frac{\tilde{\rho}_s^2}{\Delta^2 N_0^4}$$

Finite-disorder spin glass is non-coplanar

While texture is coplanar for single bond defect ($\Delta \rightarrow 0$),
state at finite Δ is non-coplanar

Histogram of scalar spin chirality per triangle



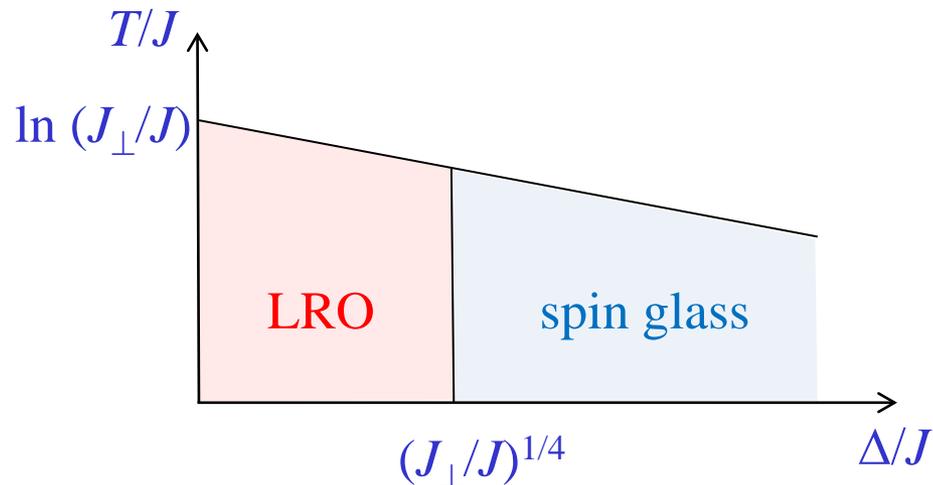
Layered triangular AFM with bond disorder

Assume weak interlayer coupling $\varepsilon \ll 1$ ($\varepsilon \sim J_{\perp}/J_{\parallel}$)

$$\chi^{\parallel}(\vec{q}, q_{\perp}) = \frac{N_0^2}{\rho_s} \frac{1}{q^2 + \varepsilon q_{\perp}^2}$$

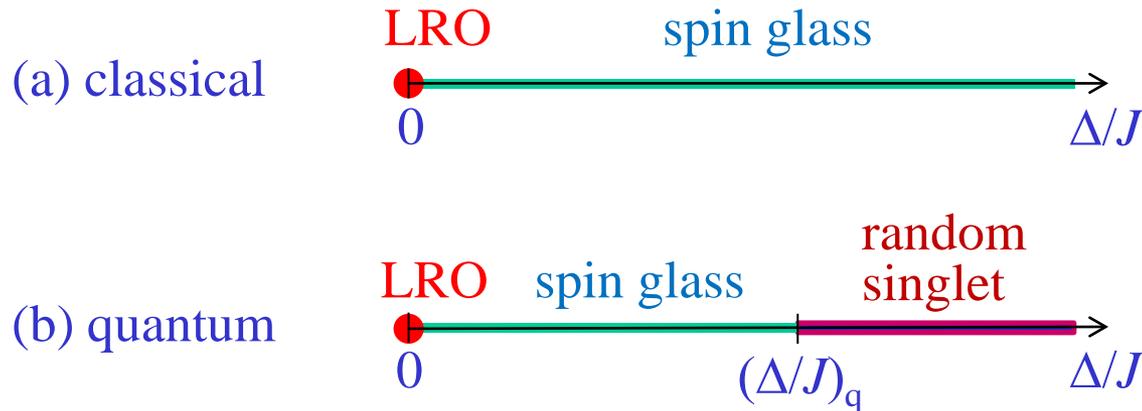
LRO is stable for small disorder, specifically if

$$\varepsilon \gtrsim \frac{\Delta^4 N_0^8}{(\tilde{\rho}_s^{\parallel})^4}$$



Summary of our results:

Phase diagram of bond-disordered triangular AF



Dey / Andrade / Vojta, arXiv:1907.08208
see also Watanabe *et al.*, JPSJ **83**, 034714 (2014)
Shimokawa *et al.*, PRB **92**, 134407 (2015)
Wu / Gong / Sheng, PRB **99**, 085141 (2019)

Note 1: Small concentration of random **vacancies** does **not** destroy LRO!

see also Maryasin/Zhitomirsky, PRB **90**, 094412 (2014)

Note 2: Easy-plane version w/ bond disorder leads to quasi-LRO!

Dey et al., in preparation

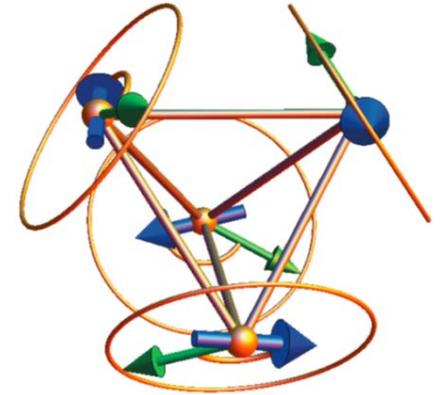
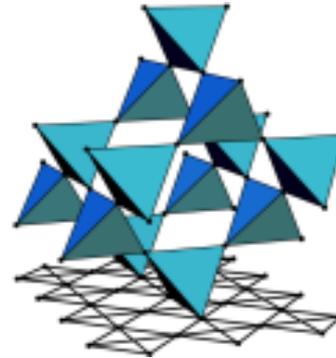
Pyrochlore *XY* antiferromagnet

+

quenched disorder

Pyrochlore XY antiferromagnet

XY moments on pyrochlore lattice
(XY planes local!)

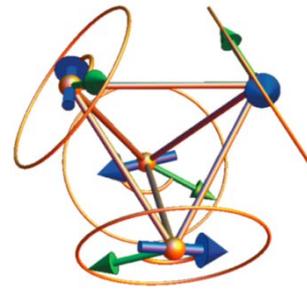


Hamiltonian
in local frame:

$$\mathcal{H} = \sum_{\langle ij \rangle} \left\{ \cancel{J_{zz} S_i^z S_j^z} - J_{\pm} (S_i^+ S_j^- + S_i^- S_j^+) \right. \\ \left. + J_{\pm\pm} (e^{i\theta_{ij}} S_i^+ S_j^+ + e^{-i\theta_{ij}} S_i^- S_j^-) \right. \\ \left. - \cancel{J_{z\pm} [S_j^z (e^{-i\theta_{ij}} S_i^+ + e^{i\theta_{ij}} S_i^-) + i \langle \rightarrow j \rangle]} \right\}$$

„XY“ model: $J_{zz} = J_{z\pm} = 0$

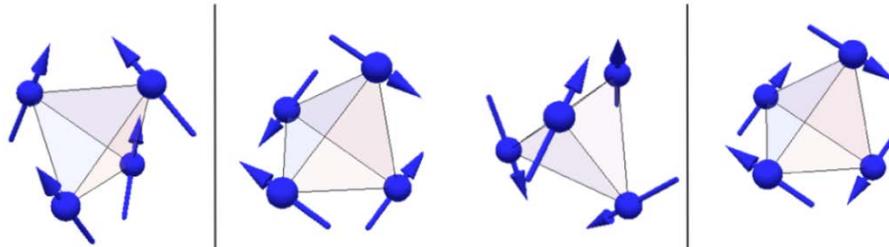
Pyrochlore XY antiferromagnet



Hamiltonian
in local frame:

$$\mathcal{H} = \sum_{\langle ij \rangle} \left\{ \cancel{J_{zz} S_i^z S_j^z} - J_{\pm} (S_i^+ S_j^- + S_i^- S_j^+) \right. \\ \left. + J_{\pm\pm} (e^{i\theta_{ij}} S_i^+ S_j^+ + e^{-i\theta_{ij}} S_i^- S_j^-) \right. \\ \left. - \cancel{J_{z\pm} [S_j^z (e^{-i\theta_{ij}} S_i^+ + e^{i\theta_{ij}} S_i^-) + i \langle \rightarrow \leftarrow \rangle_j]} \right\}$$

Classical
phase diagram



Confirmed experimental realization (with ψ_2 state):



One-parameter manifold of $Q=0$ ground states

Fluctuations select ψ_2 or ψ_3 state (both 6-fold degenerate)

(order by disorder)

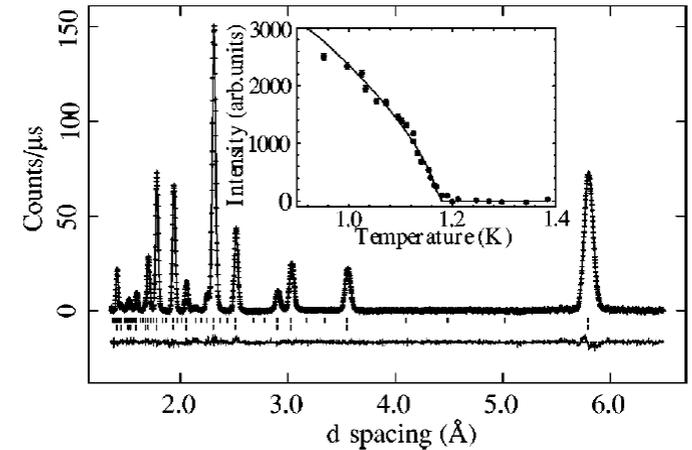
Er₂Ti₂O₇

Curie-Weiss temperature $\theta_{CW} = -13$ K

Magnetic order below $T_N = 1.2$ K

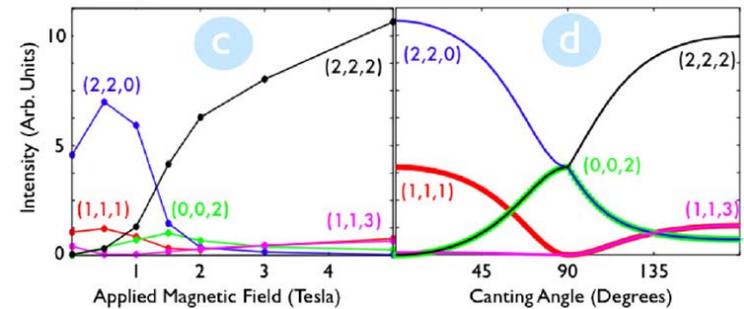
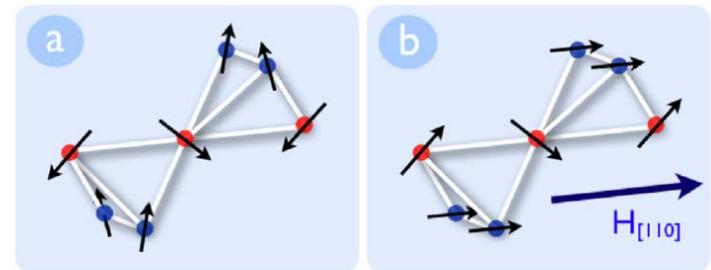
Ordering wavevector $Q = 0$

Ordered moment $\sim 3 \mu_B$



Champion *et al.*, PRB **68**, 020401 (2003)

Characteristic field dependence of Bragg peaks
as evidence for ψ_2 order



Ruff *et al.*, PRL **101**, 147205 (2008)

Spin-wave gap $\Delta \sim 0.05$ meV
consistent w/ **order-by-disorder** theory (pseudo-Goldstone)

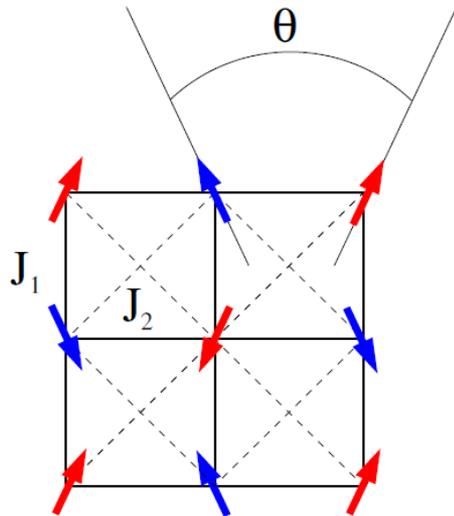
Ross *et al.*, PRL **112**, 057201 (2014)

Order by quenched disorder

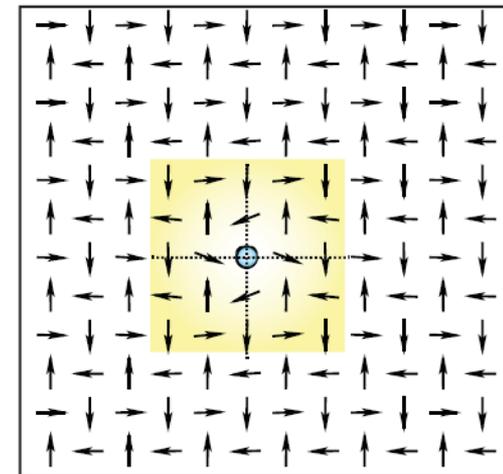
Randomness tends to select state from classically degenerate manifold
„opposite“ to that selected by thermal or quantum fluctuations (!)

Example: J_1 - J_2 square-lattice antiferromagnet

$J_2 > J_1/2$: classical energy independent of θ



Thermal and quantum fluctuations
select collinear order ($\theta=0,\pi$)



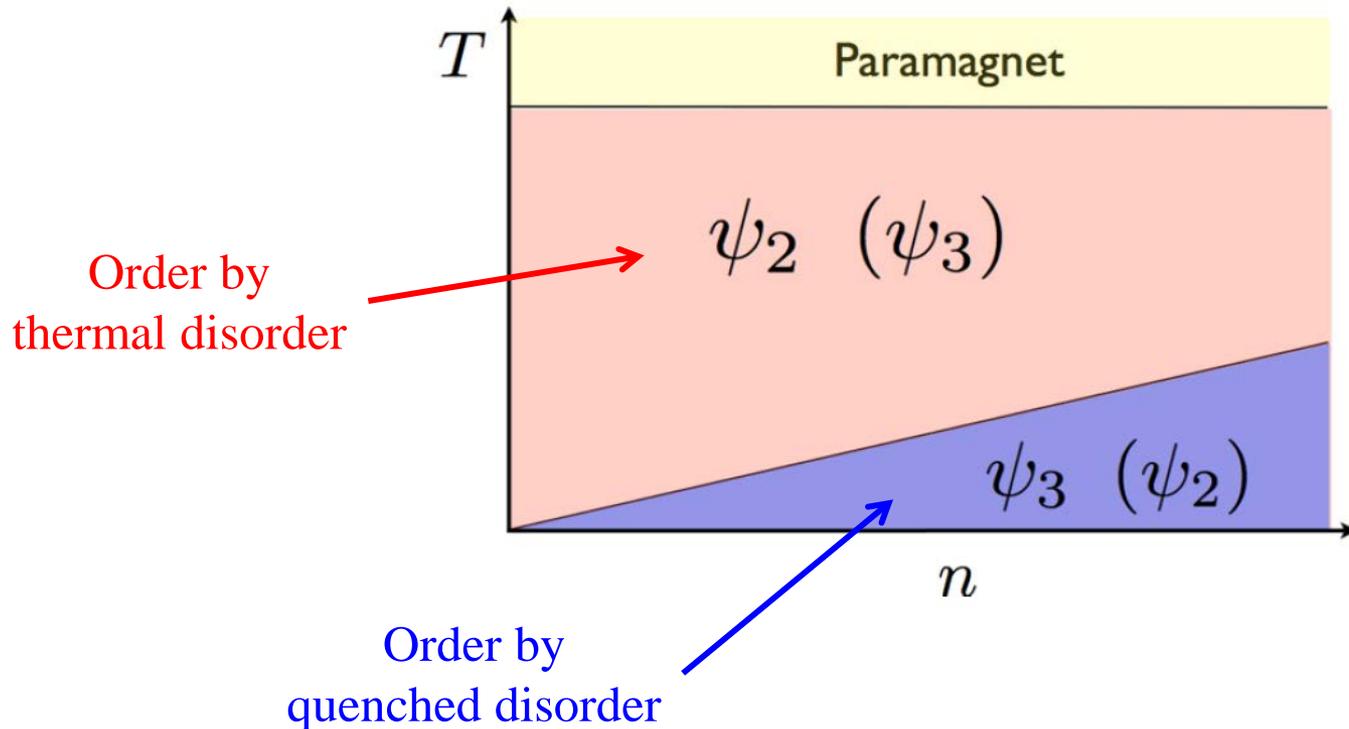
● nonmagnetic impurity

Vacancy selects
anticollinear order ($\theta=\pm\pi/2$)

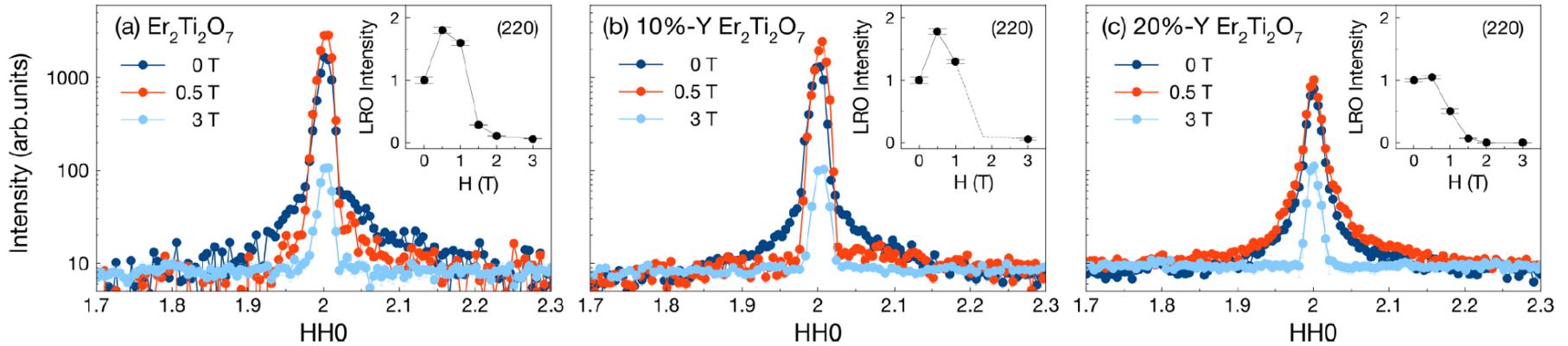
Quenched disorder in XY pyrchlores

Randomness tends to select state from classically degenerate manifold „**opposite**“ to that selected by thermal or quantum fluctuations

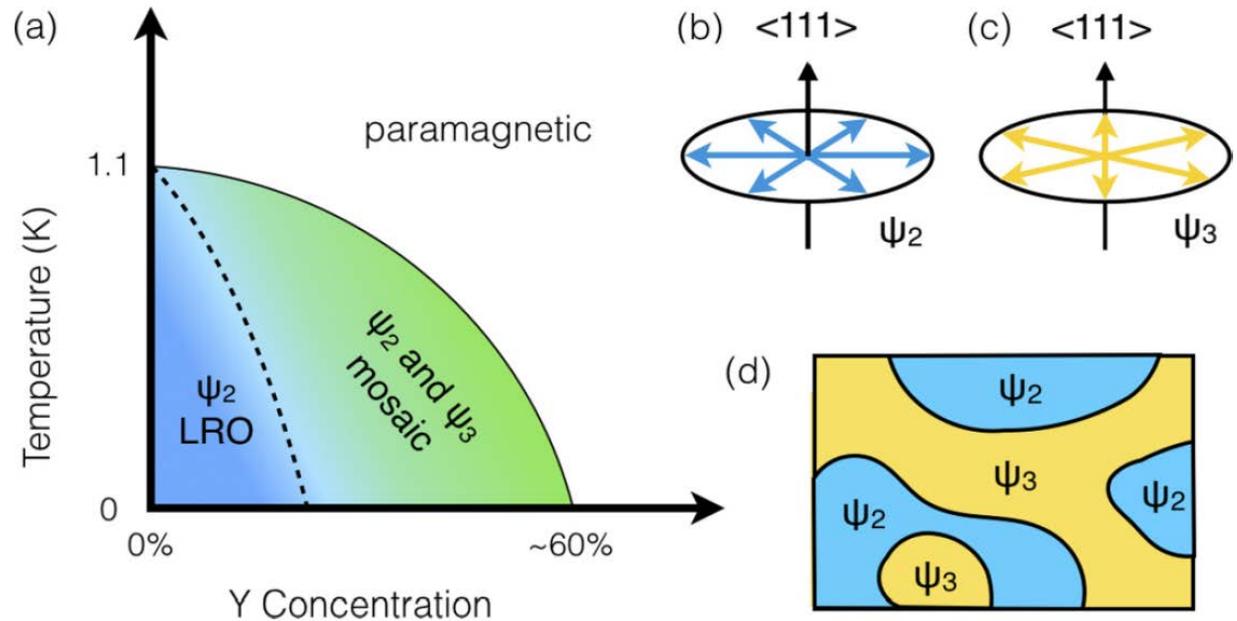
Conjectured phase diagram for classical model:



$\text{Er}_{2-x}\text{Y}_x\text{Ti}_2\text{O}_7$



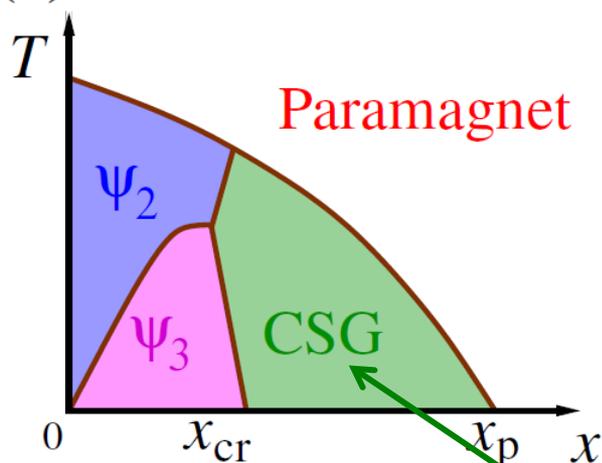
No ψ_3 LRO observed upon site dilution



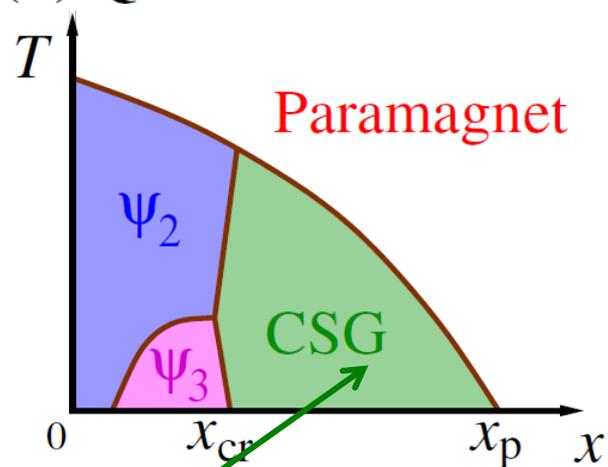
Summary of our results:

Phase diagram of dirty XY pyrochlore AF

(a) Classical

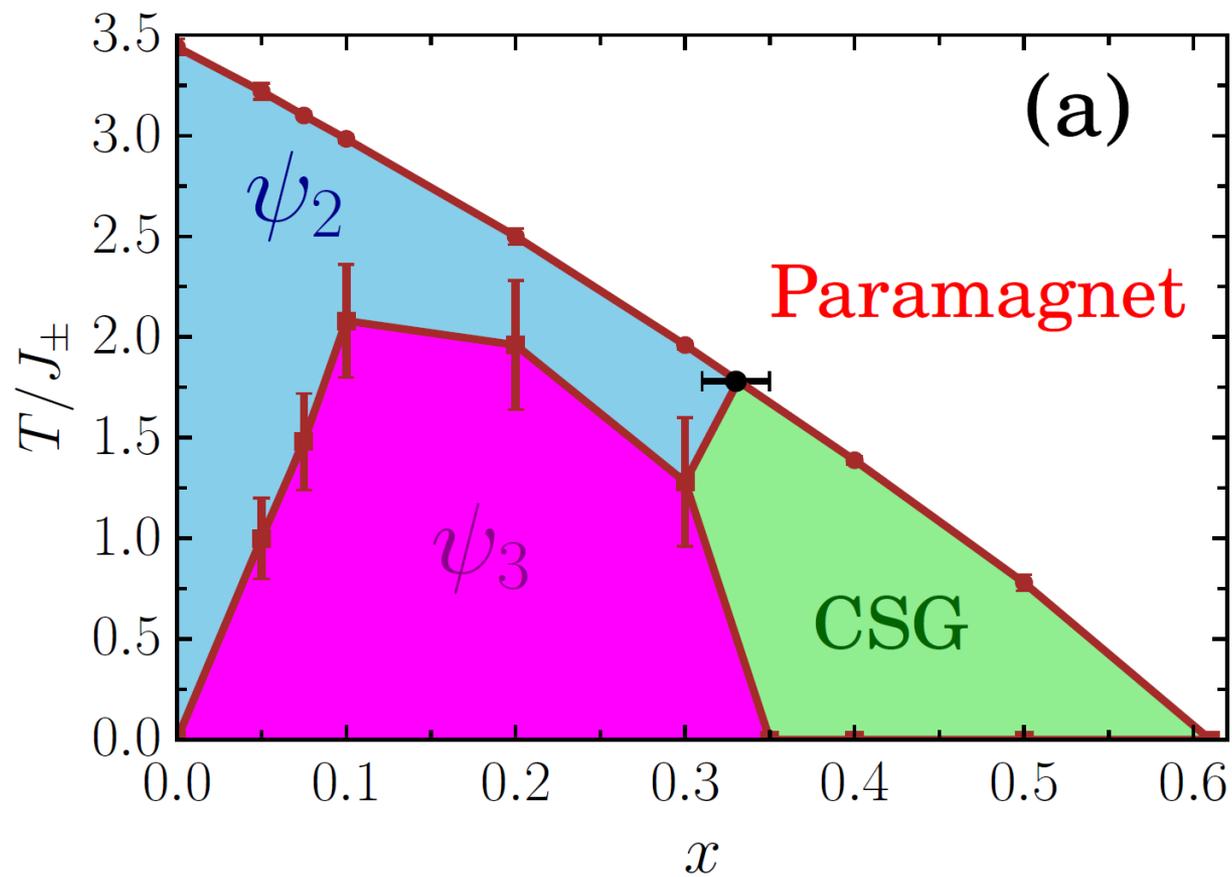


(b) Quantum



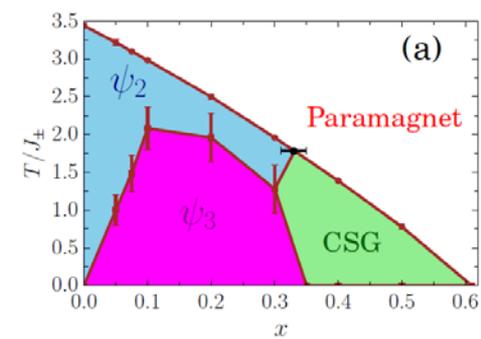
Site dilution and bond disorder
produce cluster spin glass phase

Monte Carlo results (classical): Site dilution

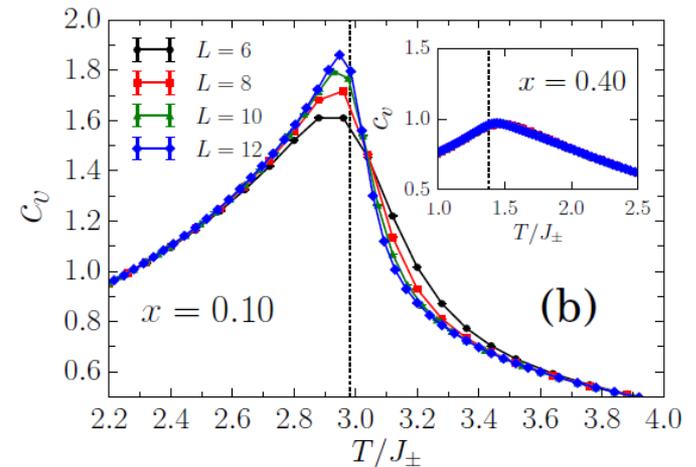


$$J_{\pm\pm}/J_{\pm} = 1$$

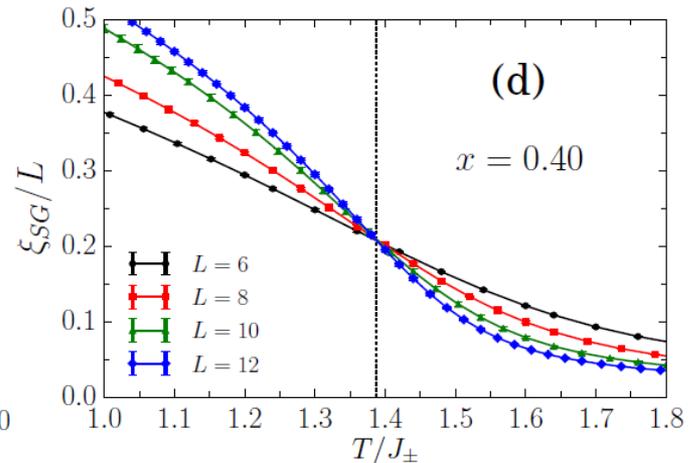
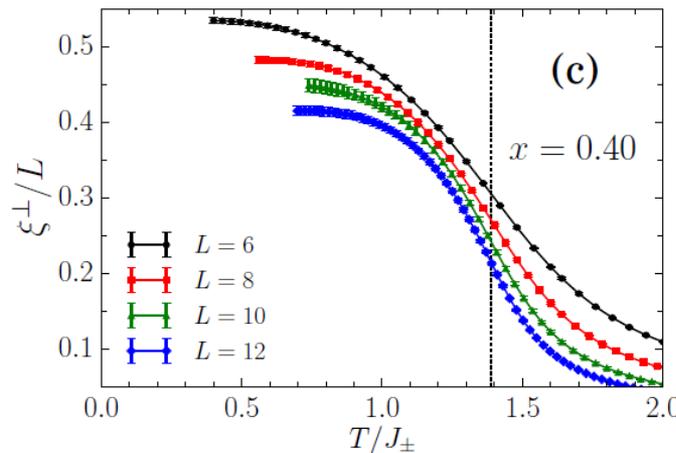
Monte Carlo results: Glassiness



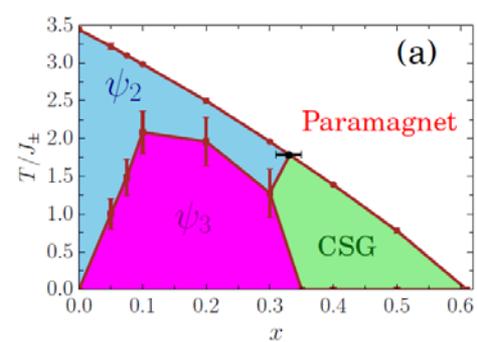
Specific heat displays sharp anomaly at $x = 0.1$ but not at $x = 0.4$



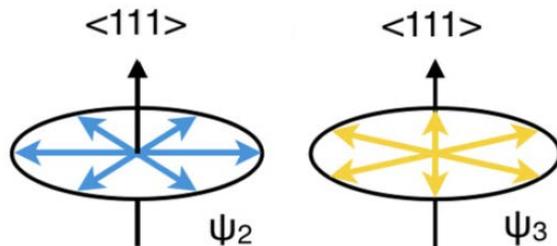
Magnetic correlation length does not display crossing points at $x = 0.4$ (while spin-glass correlation length does)



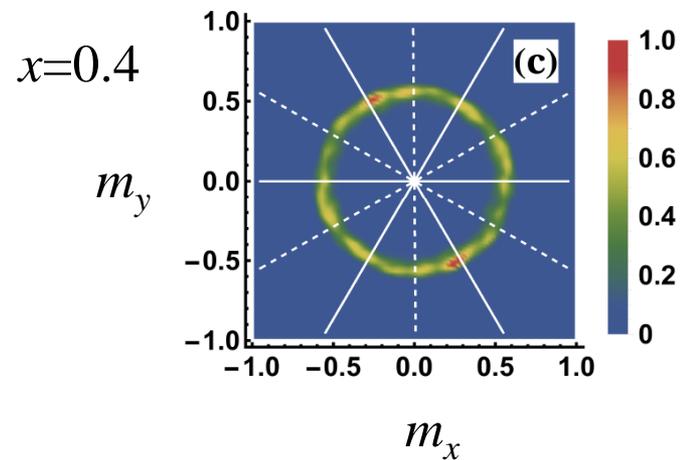
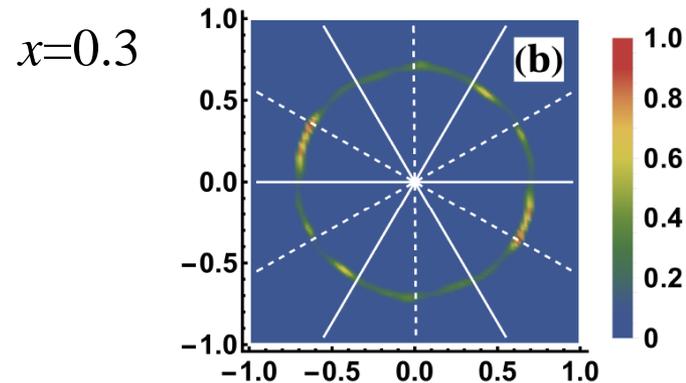
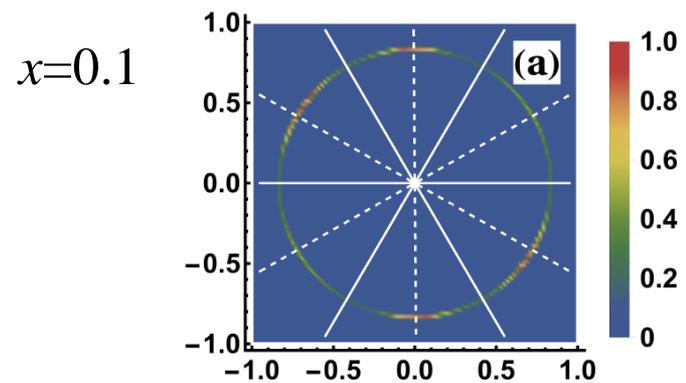
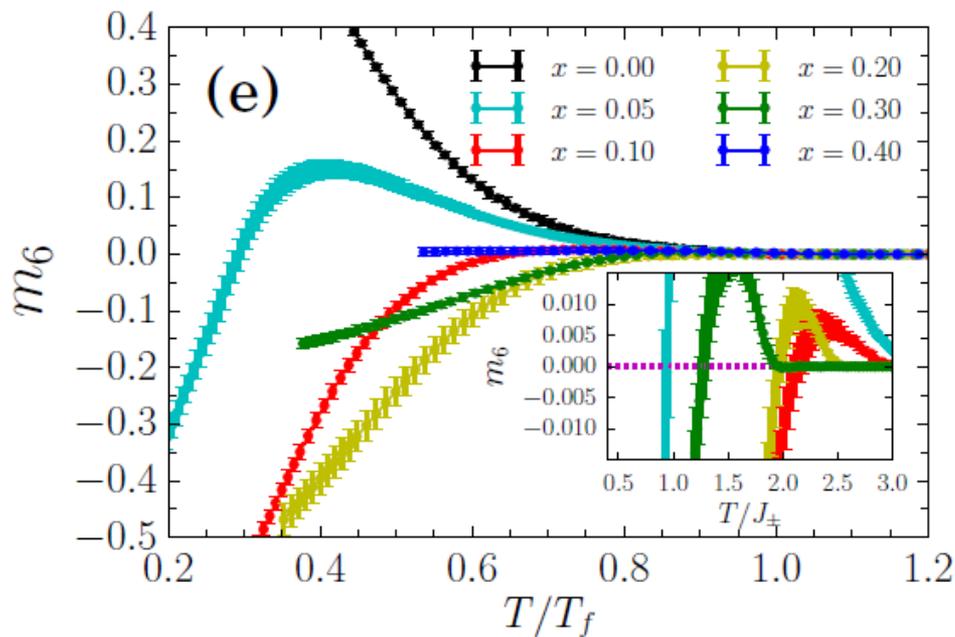
Monte Carlo results: State selection



$$m = \sqrt{m_x^2 + m_y^2}$$



$$m_6 = m \cos(6\theta) \begin{cases} > 0 \text{ for } \psi_2 \\ < 0 \text{ for } \psi_3 \end{cases}$$



Glassiness from random tranverse fields

Assume ordered state: ferromagnetic in local frame, all spins $\parallel z$

With disorder: Local mean field $\mathbf{h}_j = h_j^{\parallel} \hat{n}_{\parallel} + h_j^{\perp} \hat{n}_{\perp}$ will not be parallel to z ,
due to **off-diagonal exchange couplings**

Parameterize disorder: $J_{jk}^{\pm} = J^{\pm} (1 + \epsilon_{jk})$, $J_{jk}^{\pm\pm} = J^{\pm\pm} (1 + \epsilon_{jk})$

Effective random field $u_j = \frac{h_j^{\perp}}{J^{\pm\pm}} = \sum_{k=1}^6 \epsilon_{jk} \sin \theta_{jk}$

with strength $\delta h = \sqrt{u^2} J^{\pm\pm} = \sqrt{3x(1-x)} J^{\pm\pm}$

Transverse fluctuations
in ordered state

$$\overline{\langle S_i^{\perp 2} \rangle} = (\delta h)^2 \int \frac{d^3 q}{(2\pi)^3} \chi^{\perp}(\mathbf{q})^2$$

Ordered state stable if

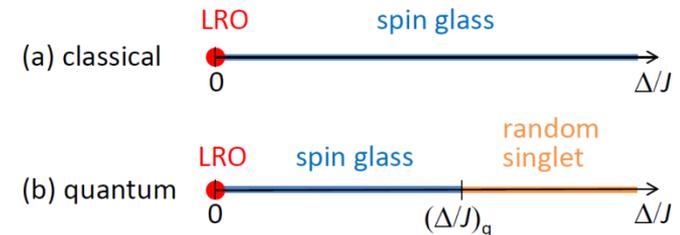
$$\delta h \ll \kappa^{d/4} \lambda^{1-d/4}$$

for bulk response given by $\chi^{\perp}(\mathbf{q}) \sim 1/(\lambda + \kappa_{\mu} q_{\mu}^2)$, i.e. gap $\Delta \propto \sqrt{\lambda}$

Summary

Quenched disorder in frustrated magnets can produce effective **random transverse fields** which destroy long-range order

1) Non-collinear LRO in triangular Heisenberg AFM is destroyed by infinitesimal quenched bond disorder in favor of **spin glass**



2) Pyrochlore XY magnets with quenched disorder show **cluster spin-glass phase** which destroys „order-by-disorder“ LRO; explains experiments in $\text{Er}_{2-x}\text{Y}_x\text{Ti}_2\text{O}_7$ and $\text{NaCaCo}_2\text{F}_7$; also applies to doped $\text{Er}_2\text{Pt}_2\text{O}_7$

