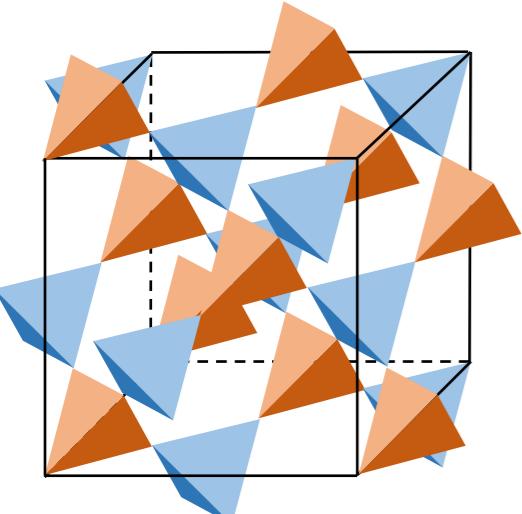


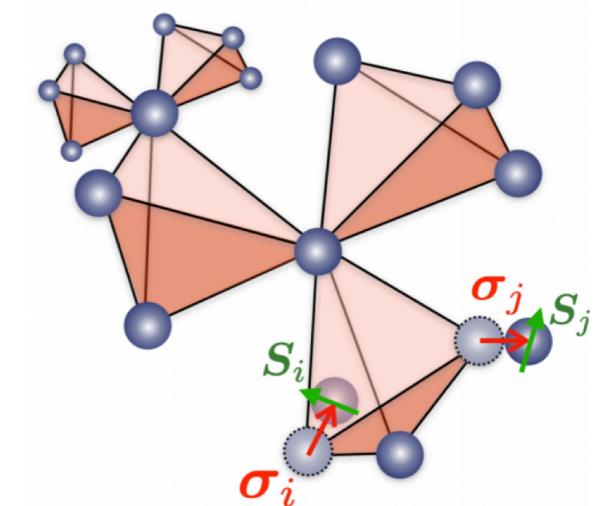
Spin-lattice glass transition without quenched disorder on pyrochlore magnet

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Coworkers
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Univ. of Tokyo, Osaka Univ.



KM, C. Hotta and H. Yoshino Phys. Rev. Lett. **124**, 087201 (2020).

KM, C. Hotta and H. Yoshino Phys. Rev. Research **4**, 033157 (2022).

KM and H. Yoshino Phys. Rev. B **107**, 054412 (2023).

Outline

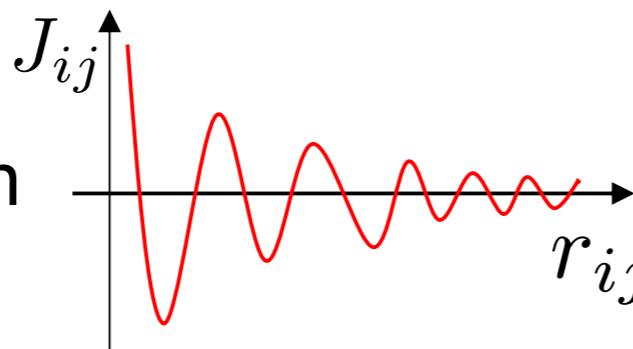
- Introduction
 - Spin glasses
 - Motivation
 - $\text{Y}_2\text{Mo}_2\text{O}_7$
- Model
- Numerical simulations (3D pyrochlore lattice)
- Mean-field theory (High dimensional limit)

Spin glasses

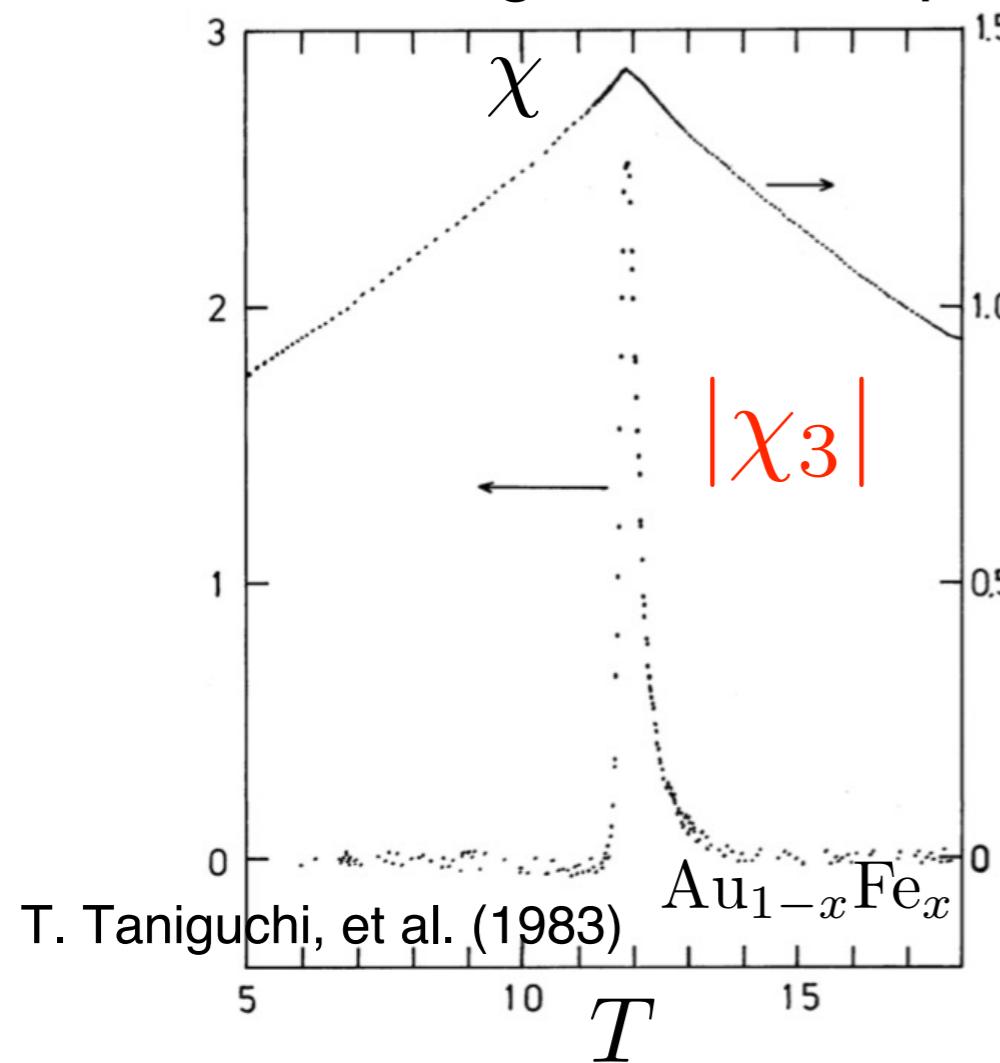
■ Canonical spin glass

$$H = \sum_{i,j} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j$$

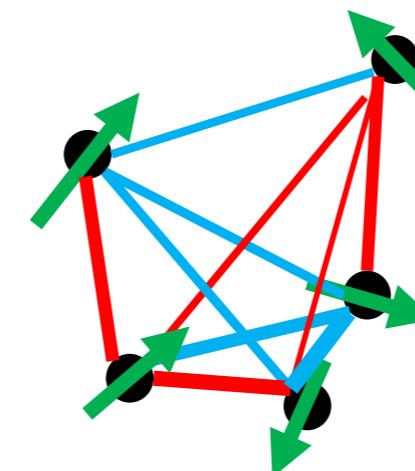
RKKY interaction



Nonlinear magnetic susceptibility



T. Taniguchi, et al. (1983)

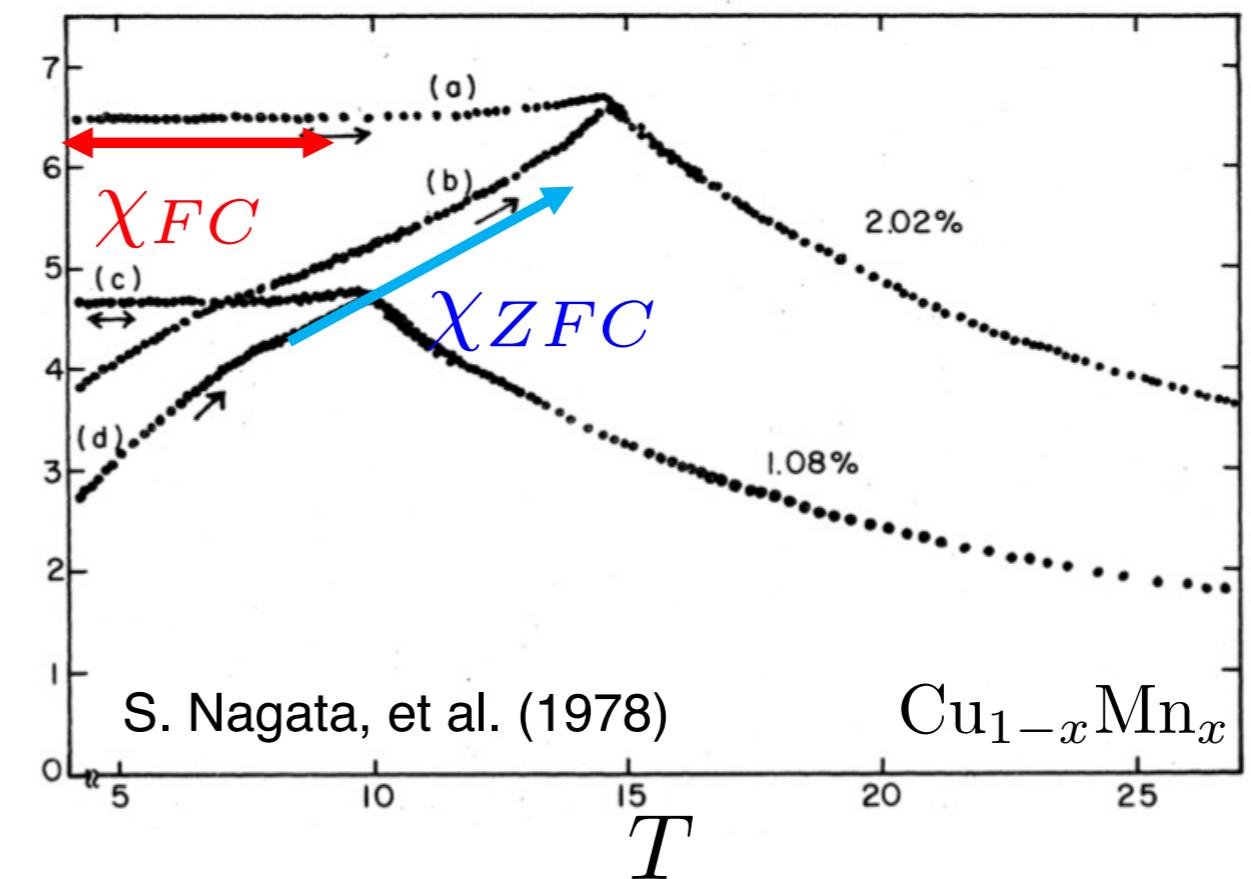


— F bond
— AF bond

Frustration due to
quenched disorder

- Thermodynamic glass transition
- Nonequilibrium in spin glass phase

FC-ZFC susceptibility



S. Nagata, et al. (1978)

Motivation

Can a spin glass transition occur without quenched disorder?

- Infinite dimension => Yes. H. Yoshino SciPost Phys. 4, 040 (2018)

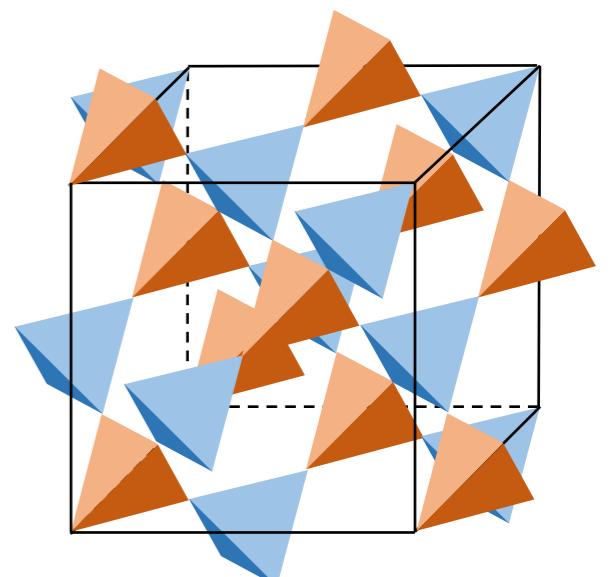
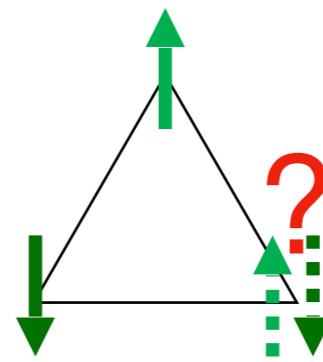
Disorder-free replica theory can be applied to spin systems.

- 3D system? ... no exact answer.

Frustration is necessary to suppress ordinary long-range order.

=> geometrically frustrated system!!

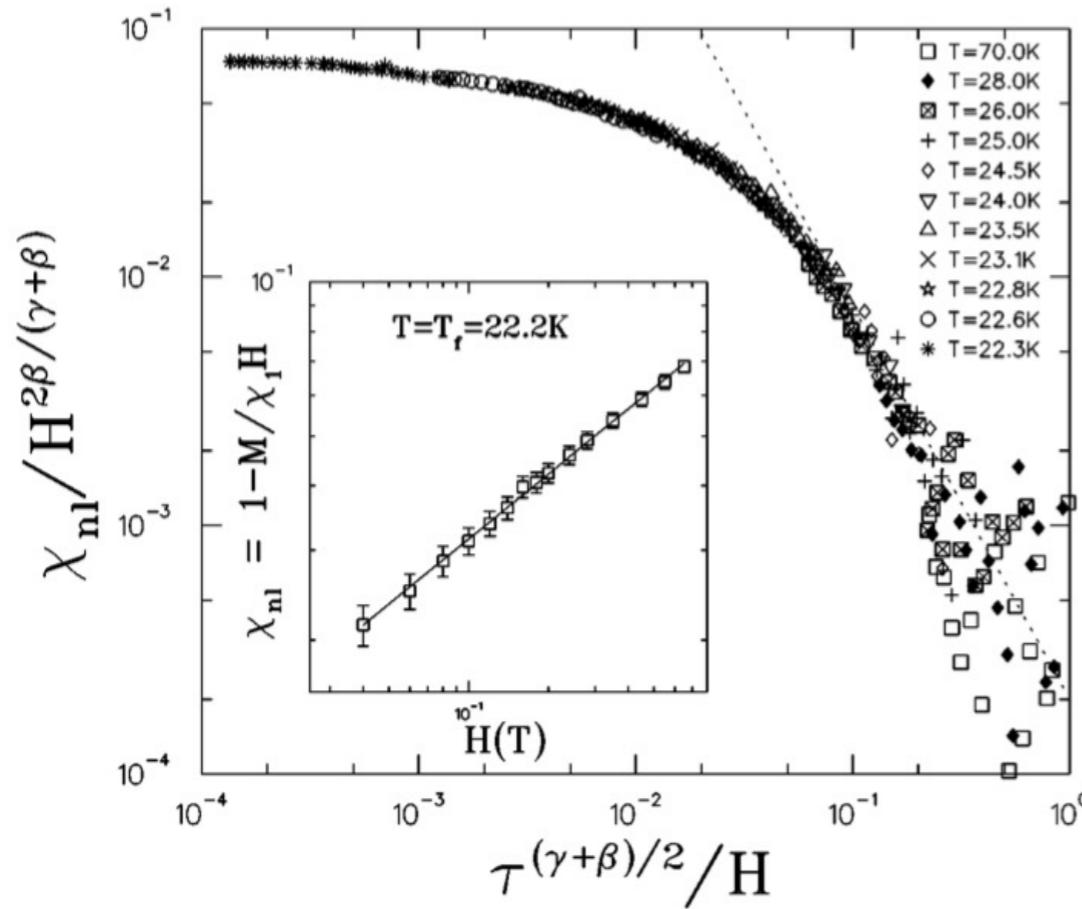
Experimental candidate $\text{Y}_2\text{Mo}_2\text{O}_7$



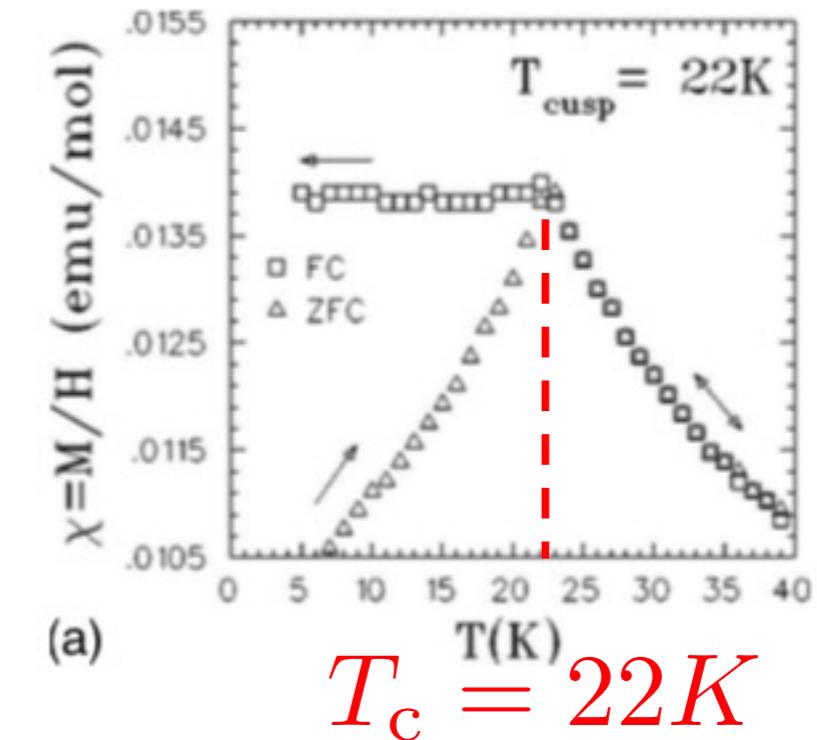


Spin-glass transition without chemical disorder

Nonlinear magnetic susceptibility



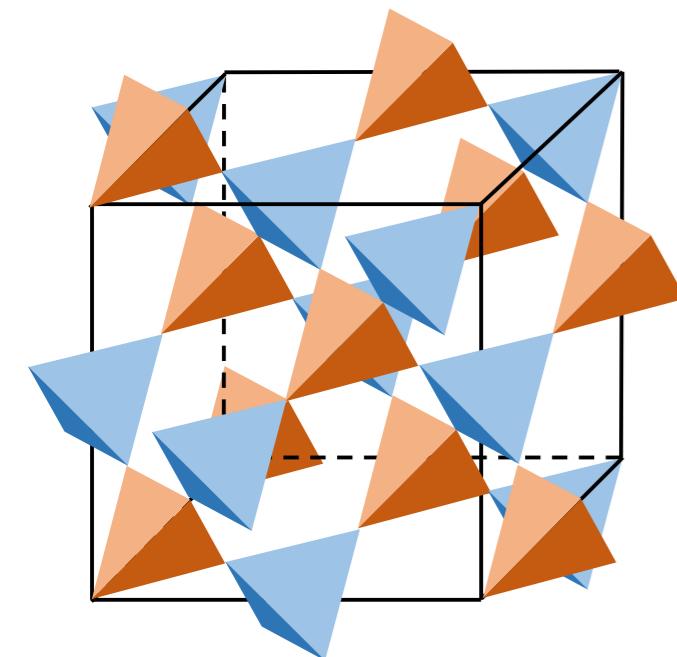
FC-ZFC susceptibility



M. J. P. Gingras et al. PRL (1997)

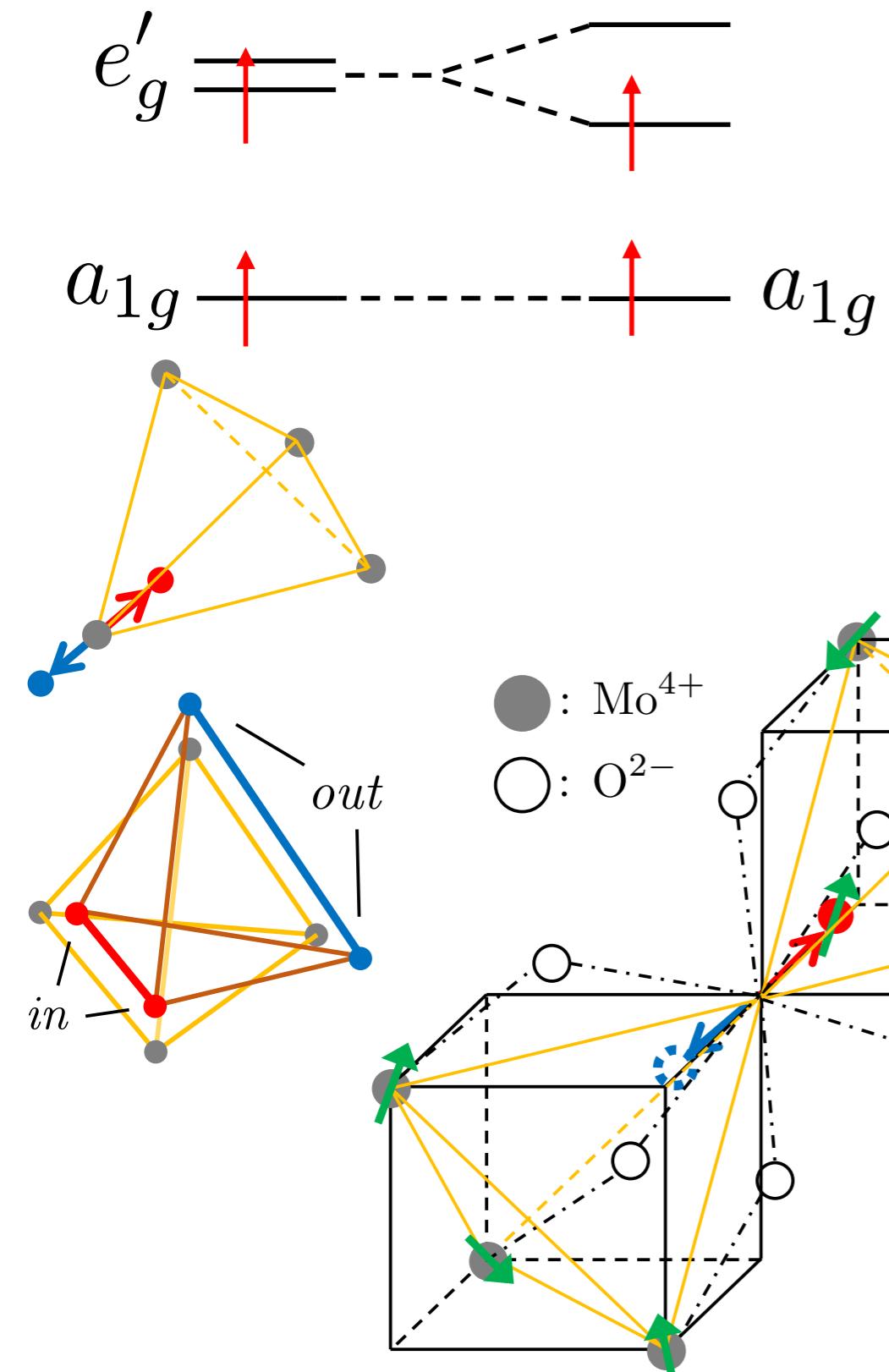
Mo⁴⁺ ($S=1$): regarded as classical Heisenberg spins around T_c

NN AF Heisenberg model on pyrochlore lattice
 \Rightarrow Classical spin liquid even at $T = 0$



What is the origin of the spin glass transition?

Lattice distortion



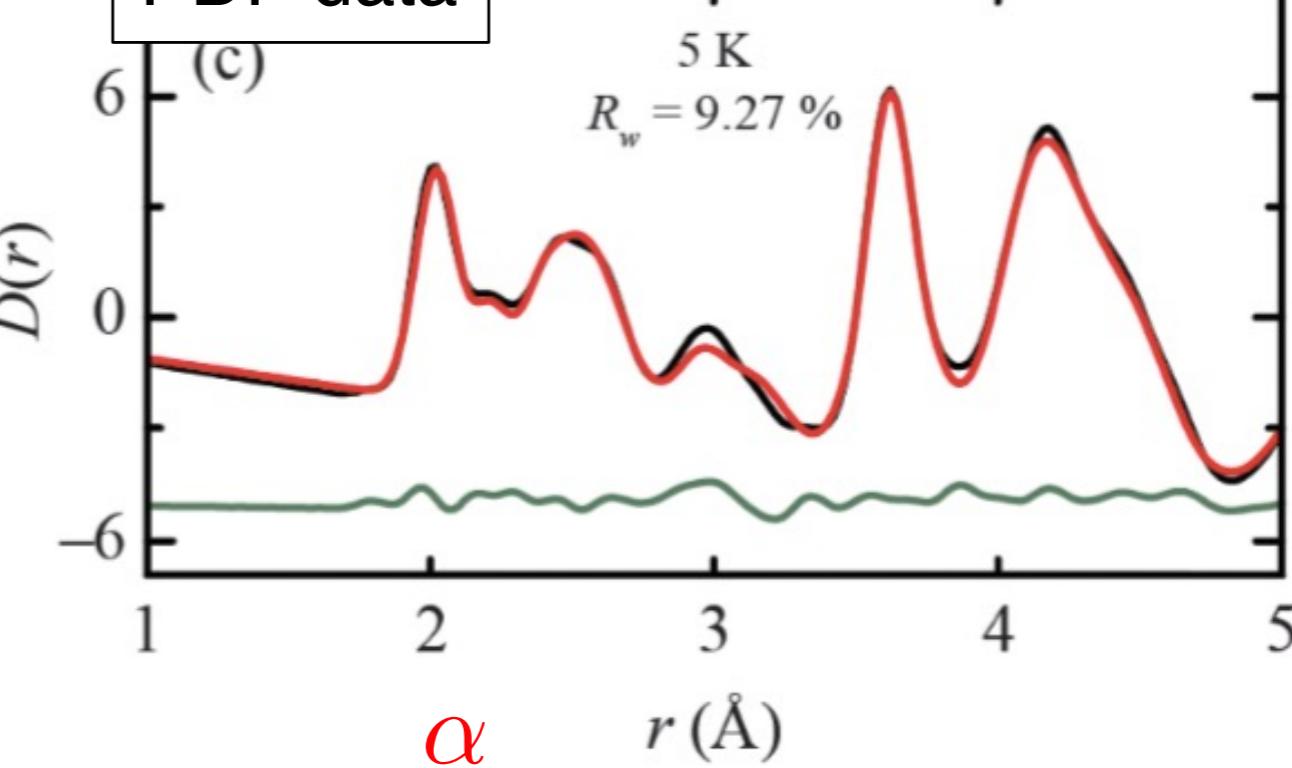
Microscopic mechanism of JT ice
=> KM, C. Hotta, H. Yoshino, PRResearch (2022)

XAFS data

C. H. Booth , et al. PRB (2000)

Atom pair	σ^2 (\AA^2)	R (\AA)	Θ_{cD} (K)	σ_{stat}^2 (\AA^2)
Mo-O(1)	0.0045(3)	2.030(2)	950(40)	0.0024(1)
Mo-Mo	0.035(1)	3.628(2)*	260(<i>fixed</i>)	0.026(5)
Mo-Y	0.0055(1)	3.628(2)*	356(20)	0.0033(4)
Y-O(1)	0.0092(3)	2.441(1)	525(12)	0.0046(3)
Y-O(2)	0.0036(1)	2.225(1)	860(60)	0.0009(3)
Y-Y/Mo	0.0042(1)	3.609(1)	381(1)	0.0020(2)

PDF data



out - out (139°) F
out - in (127°) AF
in - in (116°) AF

P.M.M. Thygesen
et al. PRL (2017)

Model

S_i : Classical Heisenberg spin $|S_i| = 1$

σ_i : Lattice displacement (orbital) $\sigma_i = \sigma_i \hat{\mathbf{e}}_\nu$ $\sigma_i = \pm 1$
 $(\nu = [111], [1\bar{1}\bar{1}], [\bar{1}1\bar{1}], [\bar{1}\bar{1}1])$

Exchange

Potential energy of distortions
 (“Spin-ice” like Hamiltonian)

$$H/J = \sum_{*j>} J_{\sigma_i, \sigma_j} S_i \cdot S_j - 3\epsilon \sum_{ij} \sigma_i \cdot \sigma_j \quad (\epsilon, J > 0)*$$

$$J_{\sigma_i, \sigma_j} = 1 + \frac{\sqrt{6}\delta}{2} (\hat{\mathbf{r}}_{ij} \cdot \sigma_i + (-\hat{\mathbf{r}}_{ij}) \cdot \sigma_i) = \begin{cases} 1 - 2\delta & (\text{out-out}) \\ 1 & (\text{in-out}) \\ 1 + 2\delta & (\text{in-in}) \end{cases}$$

$<ij>$: All nearest-neighbor pairs

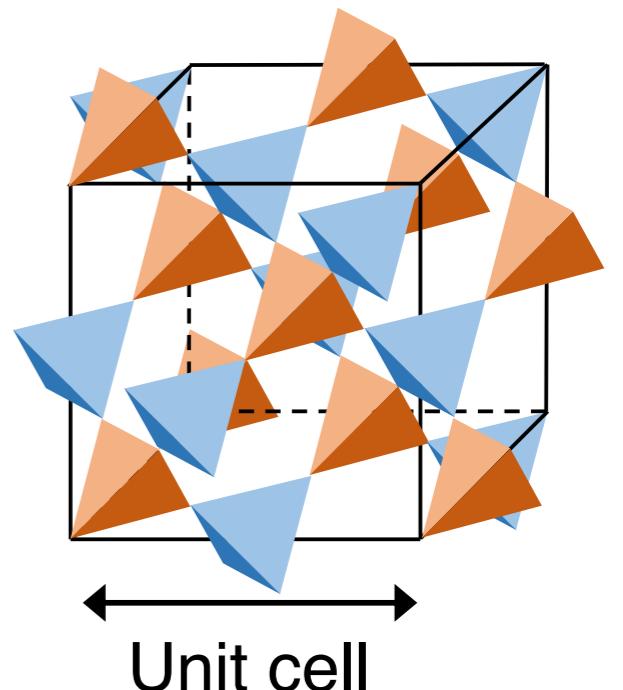
Parameter

L : System size $L = 4, 5, 6, 8$ $N = 16L^3$

δ : Amplitude $\delta = 1.5$

ϵ : Elastic energy $\epsilon = 0.6-1.2$

Method: Monte Carlo simulation



Auto-correlation function

$L = 6, \epsilon = 0.6$

Spin

$$C_S(t) = \frac{1}{N} \sum_{i=1}^N \mathbf{S}_i(0) \cdot \mathbf{S}_i(t)$$

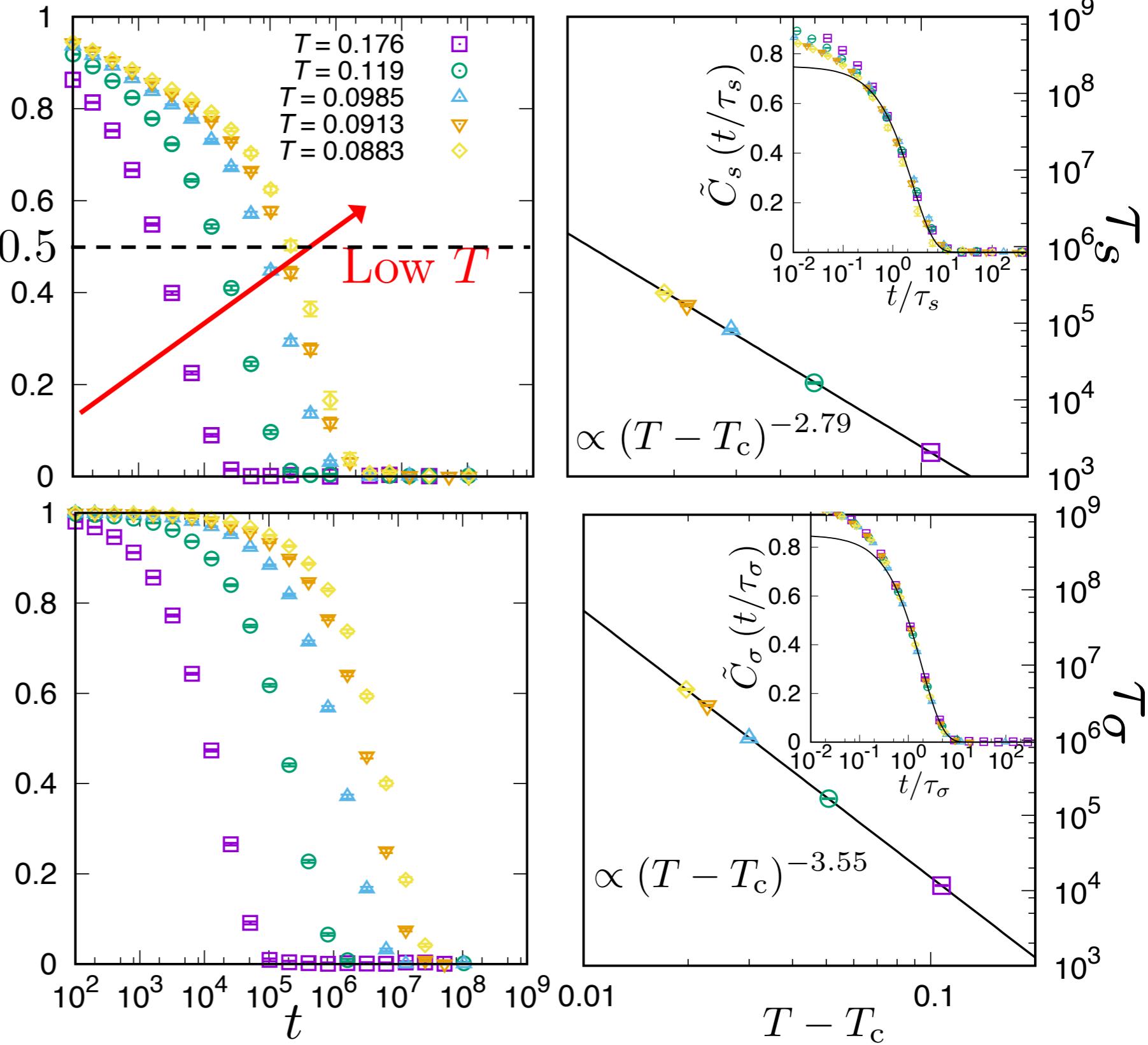
Common

$$T_c \approx 0.07$$

Lattice distortion

$$C_\sigma(t) = \frac{1}{N} \sum_{i=1}^N \sigma_i(0) \sigma_i(t)$$

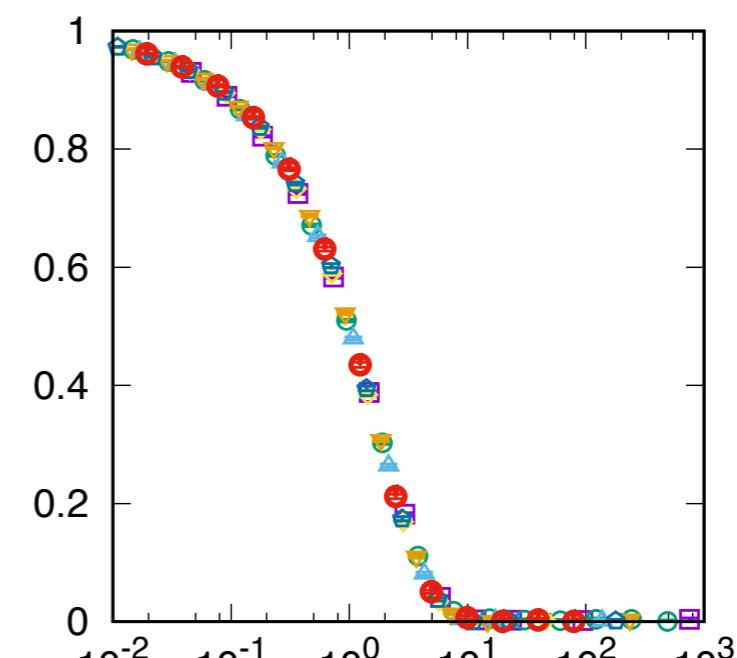
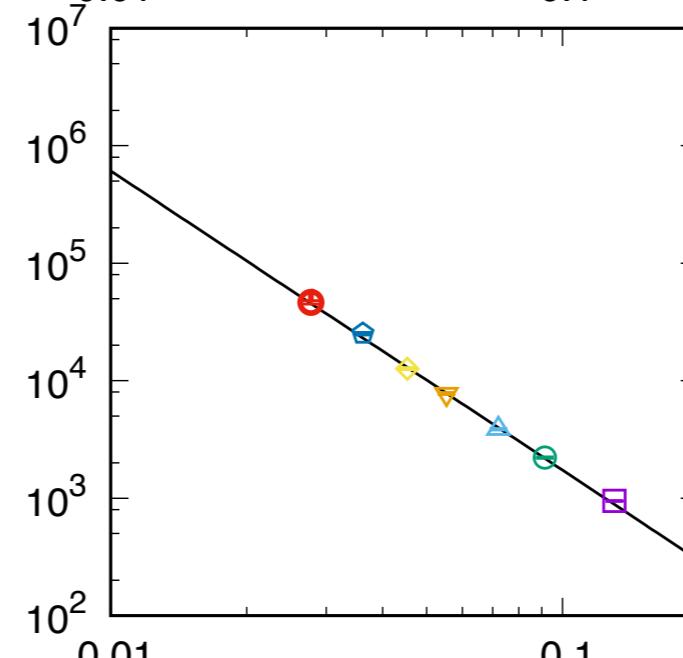
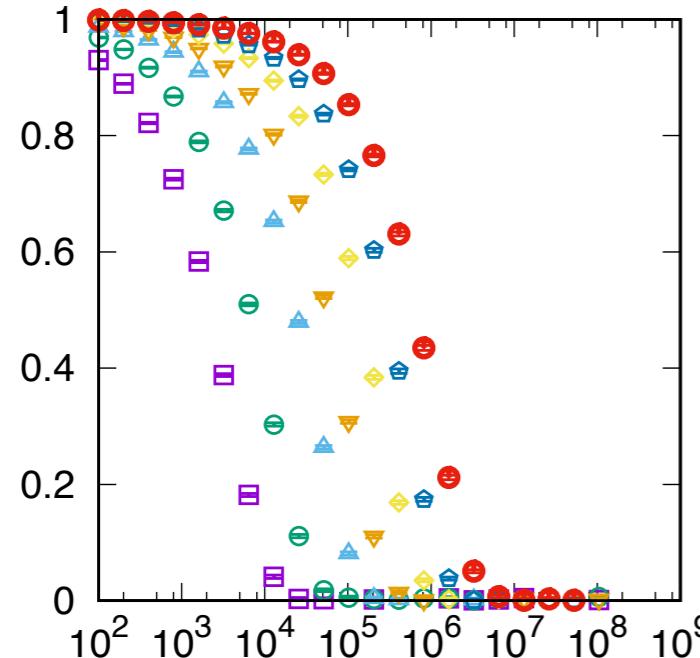
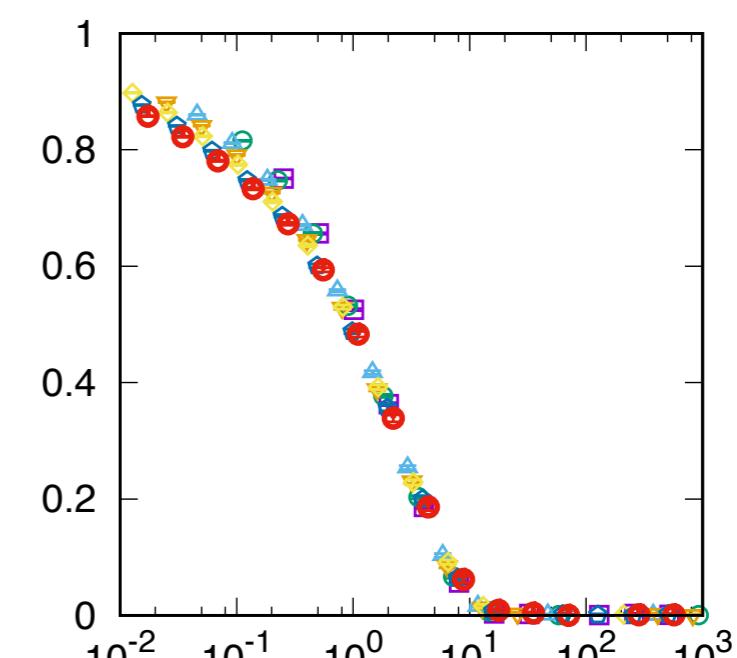
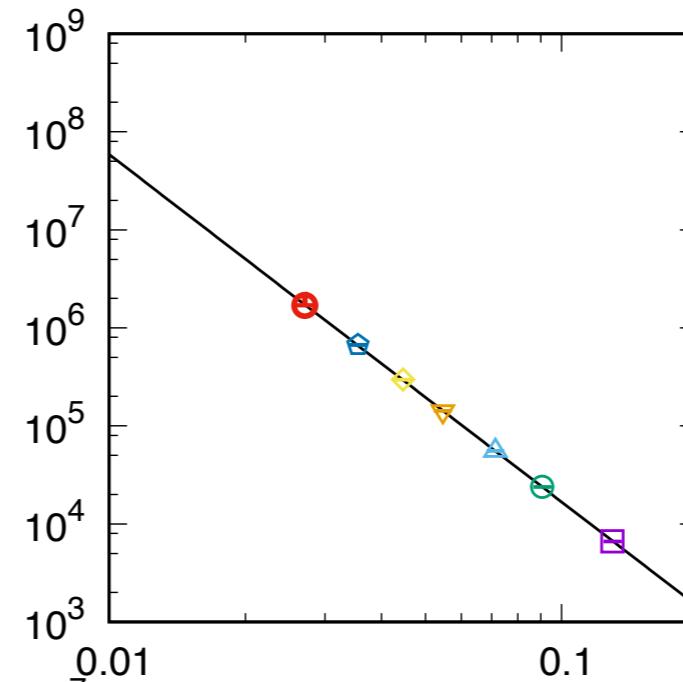
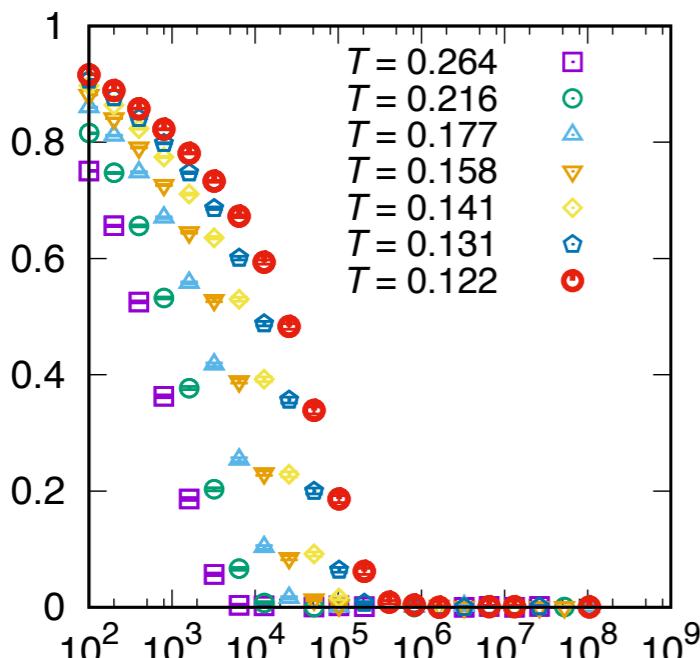
$$\tau = A(T - T_c)^{-z\nu}$$



Spin: $(T_c, z\nu) = (0.0695(19), 2.79(12))$
Lattice: $(T_c, z\nu) = (0.0686(5), 3.55(4))$

		A	T_c	$z\nu$
Spin	$\epsilon = 0.6$	3.99(89)	0.0695(19)	2.79(12)
Spin	$\epsilon = 0.65$	4.91(154)	0.0855(26)	2.55(15)
Distortion	$\epsilon = 0.6$	4.27(31)	0.0686(5)	3.55(4)
Distortion	$\epsilon = 0.65$	4.78(21)	0.0862(5)	3.54(3)

$\epsilon = 0.65, L = 5$



Structure factor ((hhI)-plane)

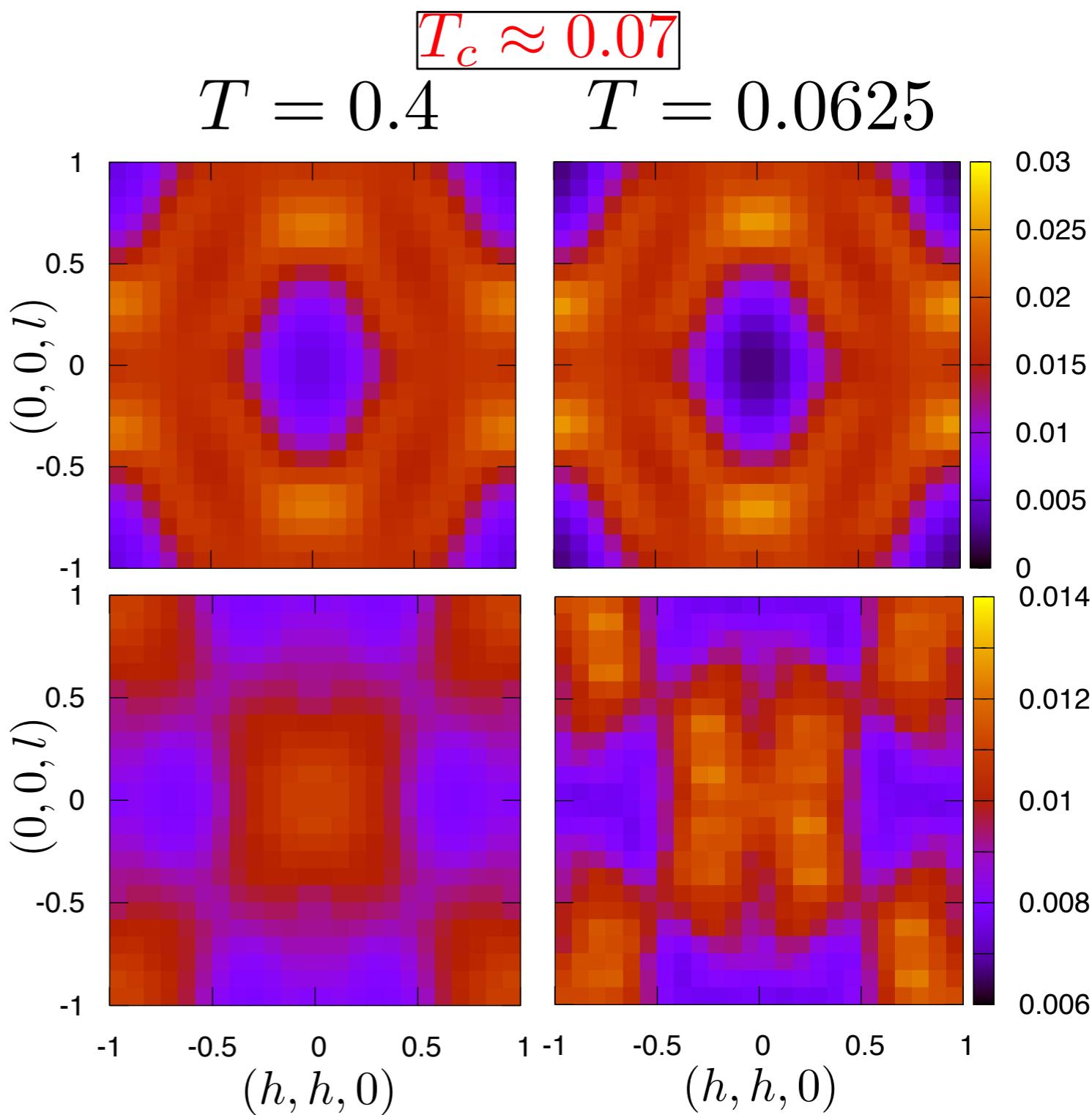
$L = 6, \epsilon = 0.6$

Spin

$$S_s(\mathbf{k}) = \frac{1}{N} \left| \sum_{i=1}^N e^{-i\mathbf{k} \cdot \mathbf{r}_i} \mathbf{S}_1 \cdot \mathbf{S}_i \right|$$

Lattice distortion

$$S_\sigma(\mathbf{k}) = \frac{1}{4N} \left| \sum_{i=1}^4 \sum_{j=1}^N e^{-i\mathbf{k} \cdot (\mathbf{r}_j - \mathbf{r}_i)} \boldsymbol{\sigma}_i \cdot \boldsymbol{\sigma}_j \right|$$



- No Brag peak at low temperature
- No notable differences between the two temperatures

→ Glass!!

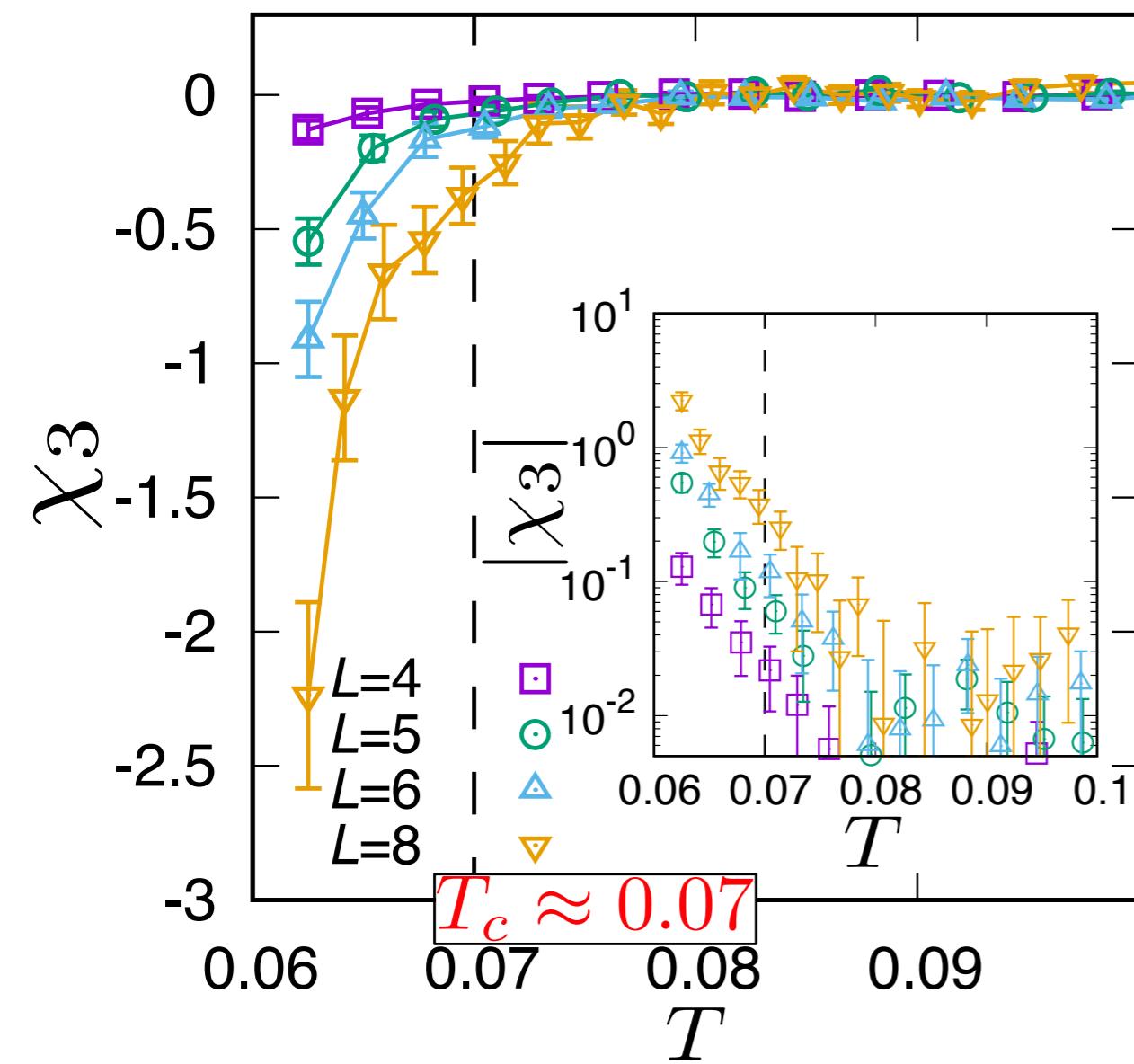
Nonlinear susceptibility

$\epsilon = 0.6$

- Magnetic ($\mu = x, y, z$)

$$m_\mu = \frac{1}{N} \sum_{i=1}^N S_{i\mu}$$

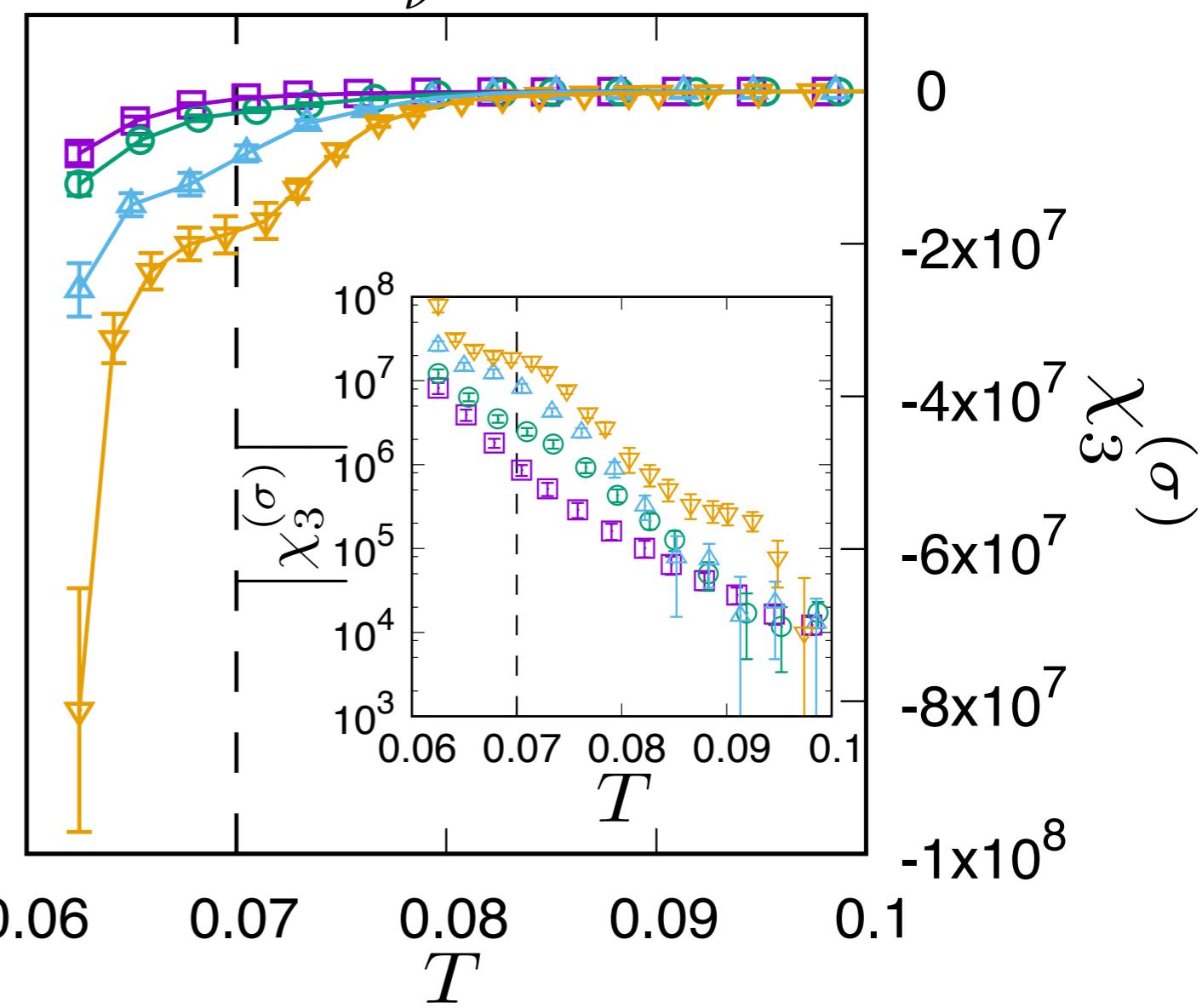
$$\chi_3 = \frac{1}{3} \sum_\mu \left. \frac{\partial^3 \langle m_\mu \rangle_{\text{eq}}}{\partial h_\mu^3} \right|_{h_\mu \rightarrow 0}$$



- Dielectric ($\nu = [111], [1\bar{1}\bar{1}], [\bar{1}1\bar{1}], [\bar{1}\bar{1}1]$)

$$p_\nu = \frac{1}{N} \sum_{i=1}^N \boldsymbol{\sigma}_i \cdot \hat{\mathbf{e}}_\nu$$

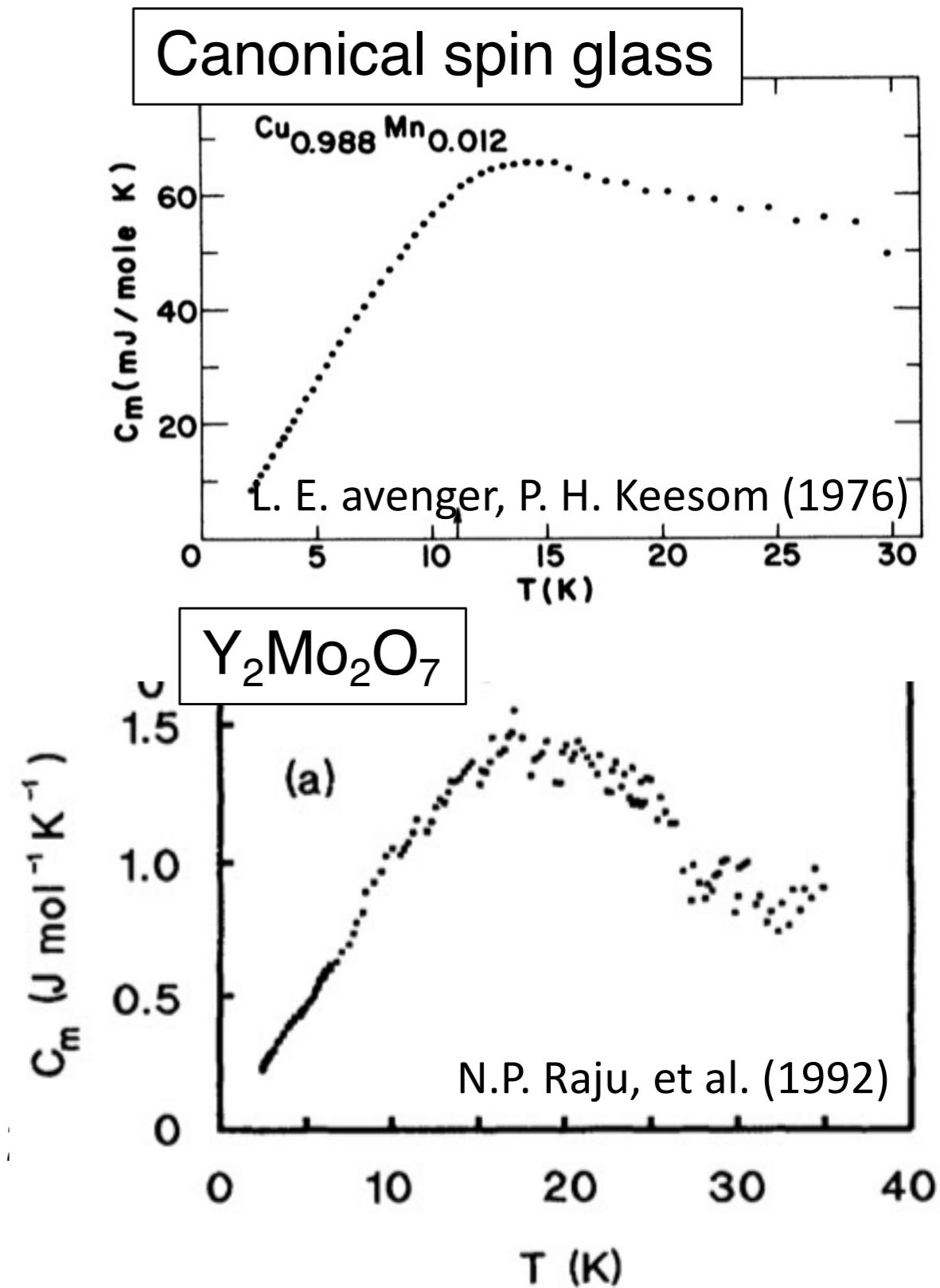
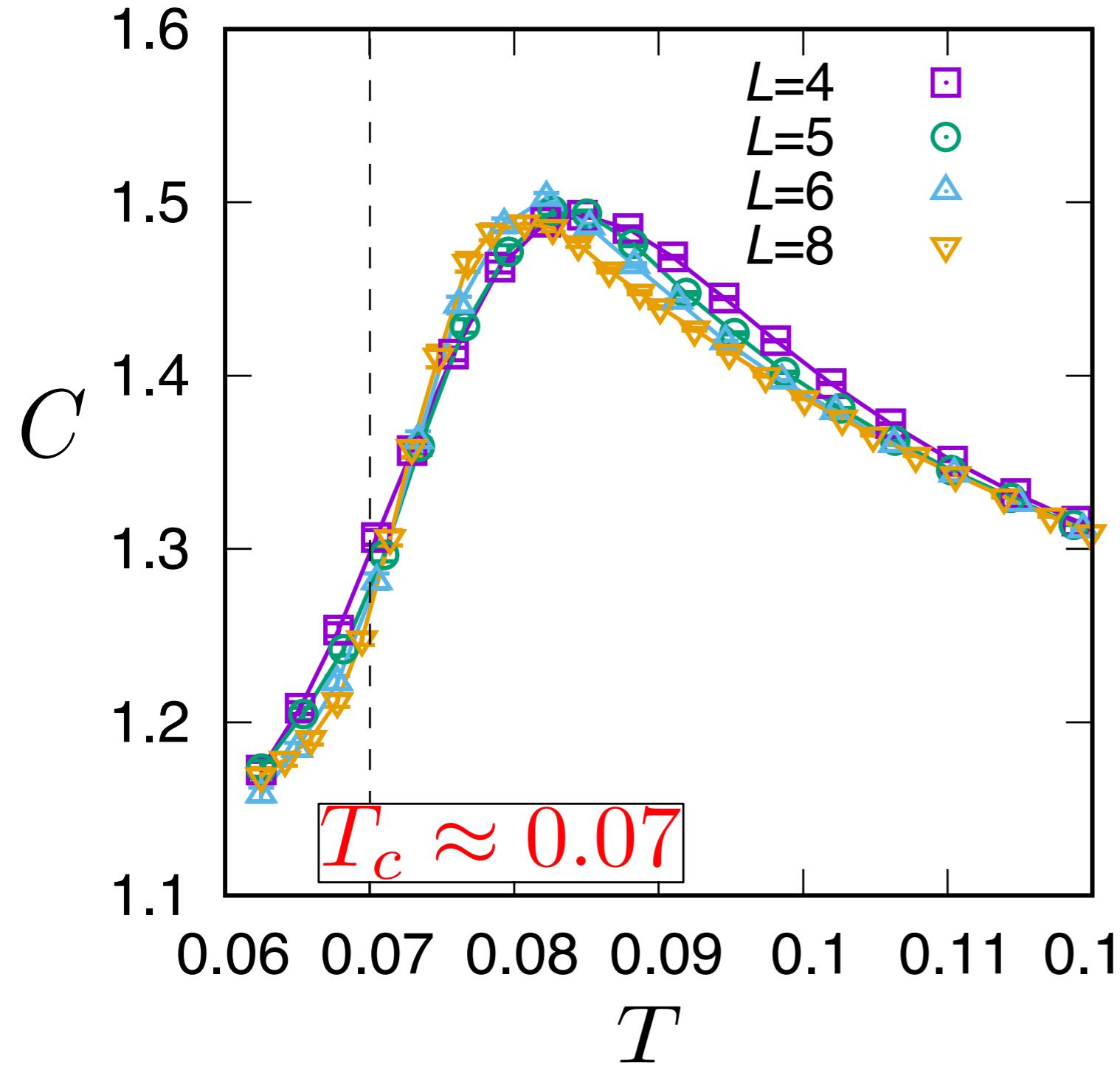
$$\chi_3^{(\sigma)} = \frac{1}{4} \sum_\nu \left. \frac{\partial^3 \langle p_\nu \rangle_{\text{eq}}}{\partial E_\nu^3} \right|_{E_\nu \rightarrow 0}$$



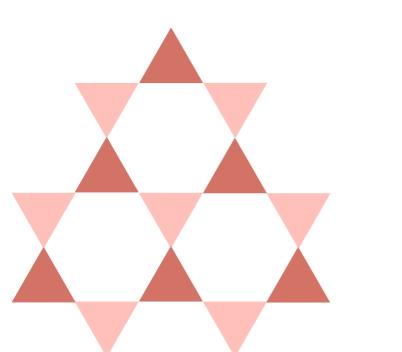
Heat capacity

$$\epsilon = 0.6$$

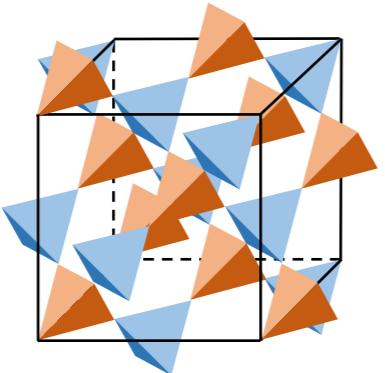
$$C = N^{-1} \beta^2 (\langle E^2 \rangle_{\text{eq}} - \langle E \rangle_{\text{eq}}^2) \quad \beta = 1/T$$



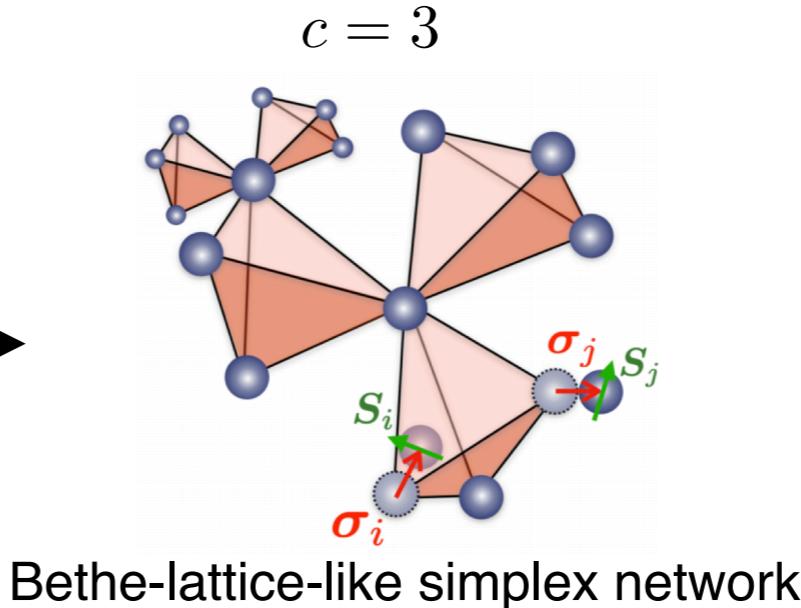
Mean-field theory



Kagome



Pyrochlore



$N \gg c \gg 1$

$$\text{with } \alpha = \frac{c}{M}$$

$$H = \frac{1}{\sqrt{c(k-1)}} \sum_{\langle i,j \rangle} [J_{\sigma_i, \sigma_j} \mathbf{S}_i \cdot \mathbf{S}_j + \epsilon \boldsymbol{\sigma}_i \cdot \boldsymbol{\sigma}_j]$$

$$J_{\sigma_i, \sigma_j} = J[1 + \delta(\boldsymbol{\sigma}_i \cdot \hat{\mathbf{r}}_{ij} + \boldsymbol{\sigma}_j \cdot \hat{\mathbf{r}}_{ji})]$$

Spherical constraint

$$\mathbf{S}_i = (S_i^1, S_i^2, \dots, S_i^M) \quad \sum_{i=1}^N |\mathbf{S}_i|^2 = NM$$

$$\boldsymbol{\sigma} = (\sigma_i^1, \sigma_i^2, \dots, \sigma_i^c) \quad \sum_{i=1}^N |\boldsymbol{\sigma}_i|^2 = Nc$$

Replica trick

$$-\beta F = \log Z = \lim_{n \rightarrow 0} \partial_n Z^n$$

$\delta = 0$ (spin-lattice decoupled):
essentially the same as the spherical SK model
=> Replica symmetry breaking doesn't occur

Order parameter

- spin

$$Q_{ab} = \lim_{N \rightarrow \infty} \frac{1}{NM} \sum_{i=1}^N \langle \mathbf{S}_i^a \cdot \mathbf{S}_i^b \rangle$$

- lattice distortion

$$q_{ab} = \lim_{N \rightarrow \infty} \frac{1}{Nc} \sum_{i=1}^N \langle \boldsymbol{\sigma}_i^a \cdot \boldsymbol{\sigma}_i^b \rangle$$

Free-energy functional

$$Z^n \propto \prod_{a \leq b} \left(\int_{-\infty}^{\infty} dQ_{ab} \int_{-\infty}^{\infty} dq_{ab} \right) e^{(Nc)s_n[\hat{Q}, \hat{q}]}$$

$$s_n[\hat{Q}, \hat{q}] = s_{\text{ent}}^{(s)}[\hat{Q}] + s_{\text{ent}}^{(\sigma)}[\hat{q}] + \mathcal{F}_{\text{int}}[\hat{Q}, \hat{q}] \quad s_{\text{ent}}^{(s)}[\hat{Q}] = \frac{1}{\alpha} \left[\frac{n}{2} [1 + \log(2\pi)] + \frac{1}{2} \log(\det \hat{Q}) \right],$$

Saddle point equations

$$0 = \frac{\partial s_n[\hat{Q}, \hat{q}]}{\partial Q_{ab}} \Big|_{\hat{Q}=\hat{Q}^*, \hat{q}=\hat{q}^*} = \frac{\partial s_n[\hat{Q}, \hat{q}]}{\partial q_{ab}} \Big|_{\hat{Q}=\hat{Q}^*, \hat{q}=\hat{q}^*}$$

$$\mathcal{F}_{\text{int}}[\hat{Q}, \hat{q}] = \frac{\beta^2}{4} \sum_{a,b} \left[\frac{J^2}{\alpha} (1 + 2\delta^2 q_{ab}) Q_{ab}^2 + \epsilon^2 q_{ab}^2 \right]$$

$$\text{free-energy functional} \quad -\beta f[\hat{Q}^*, \hat{q}^*] = \frac{-\beta F[\hat{Q}^*, \hat{q}^*]}{Nc} = \frac{\partial s_n[\hat{Q}^*, \hat{q}^*]}{\partial n} \Big|_{n=0}$$

$$\text{RS ansatz} \quad Q_{ab} = Q + (1 - Q)\delta_{ab}, \quad q_{ab} = Q + (1 - q)\delta_{ab}$$

$$\text{Hessian matrix} \quad \hat{\mathcal{H}} = \begin{bmatrix} H_{QQ} & H_{Qq} \\ H_{qQ} & H_{qq} \end{bmatrix}$$

The eigen value must be non-negative

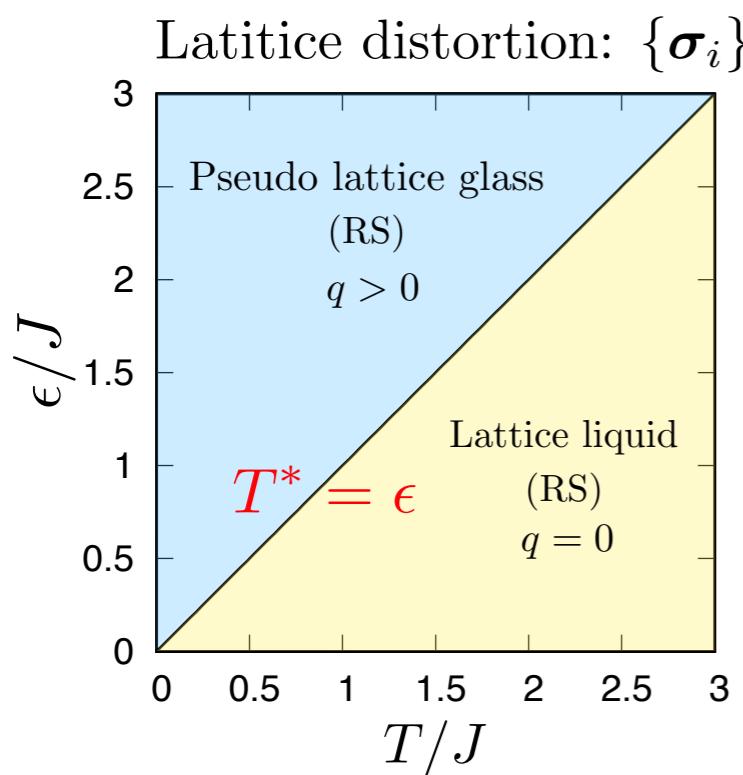
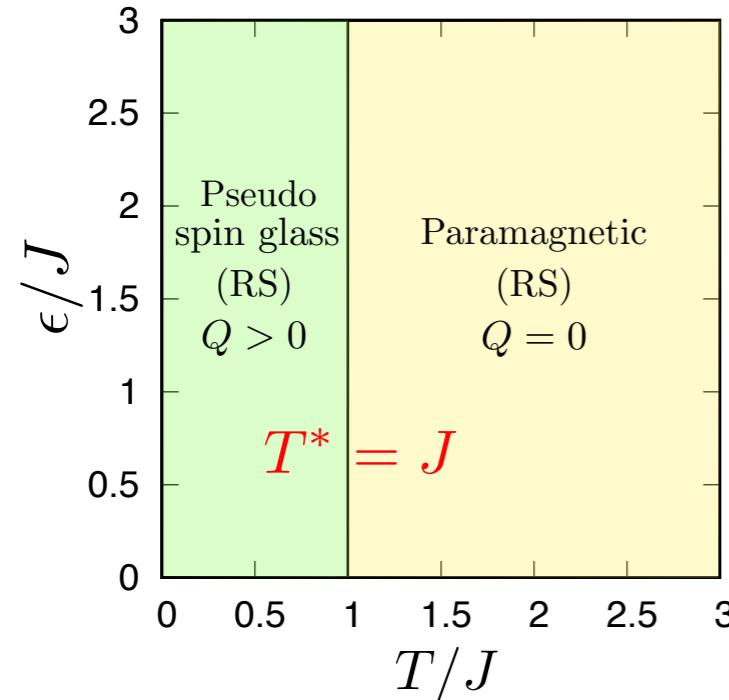
$$H_{Q_{ab}, Q_{cd}} \equiv -\frac{\partial^2 s_n[\hat{Q}, \hat{q}]}{\partial Q_{ab} \partial Q_{cd}} \quad H_{q_{ab}, q_{cd}} \equiv -\frac{\partial^2 s_n[\hat{Q}, \hat{q}]}{\partial q_{ab} \partial q_{cd}}$$

$$H_{Q_{ab}, q_{cd}} \equiv -\frac{\partial^2 s_n[\hat{Q}, \hat{q}]}{\partial Q_{ab} \partial q_{cd}}$$

Phase diagram

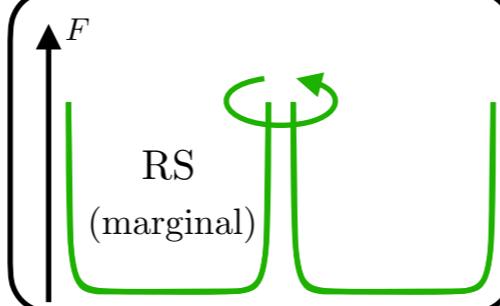
$\delta = 0$ (spin-lattice decoupled)

Spin: $\{S_i\}$

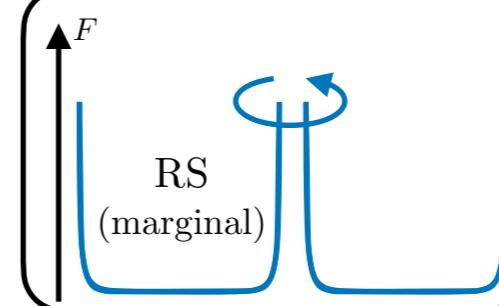


Artifact of the high dimensional limit

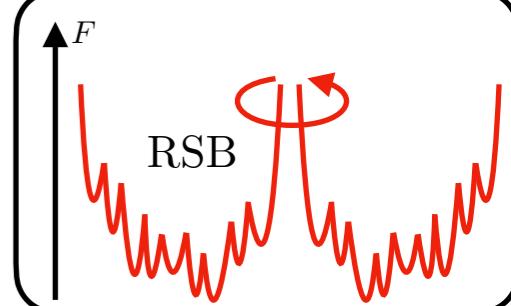
pseudo spin glass



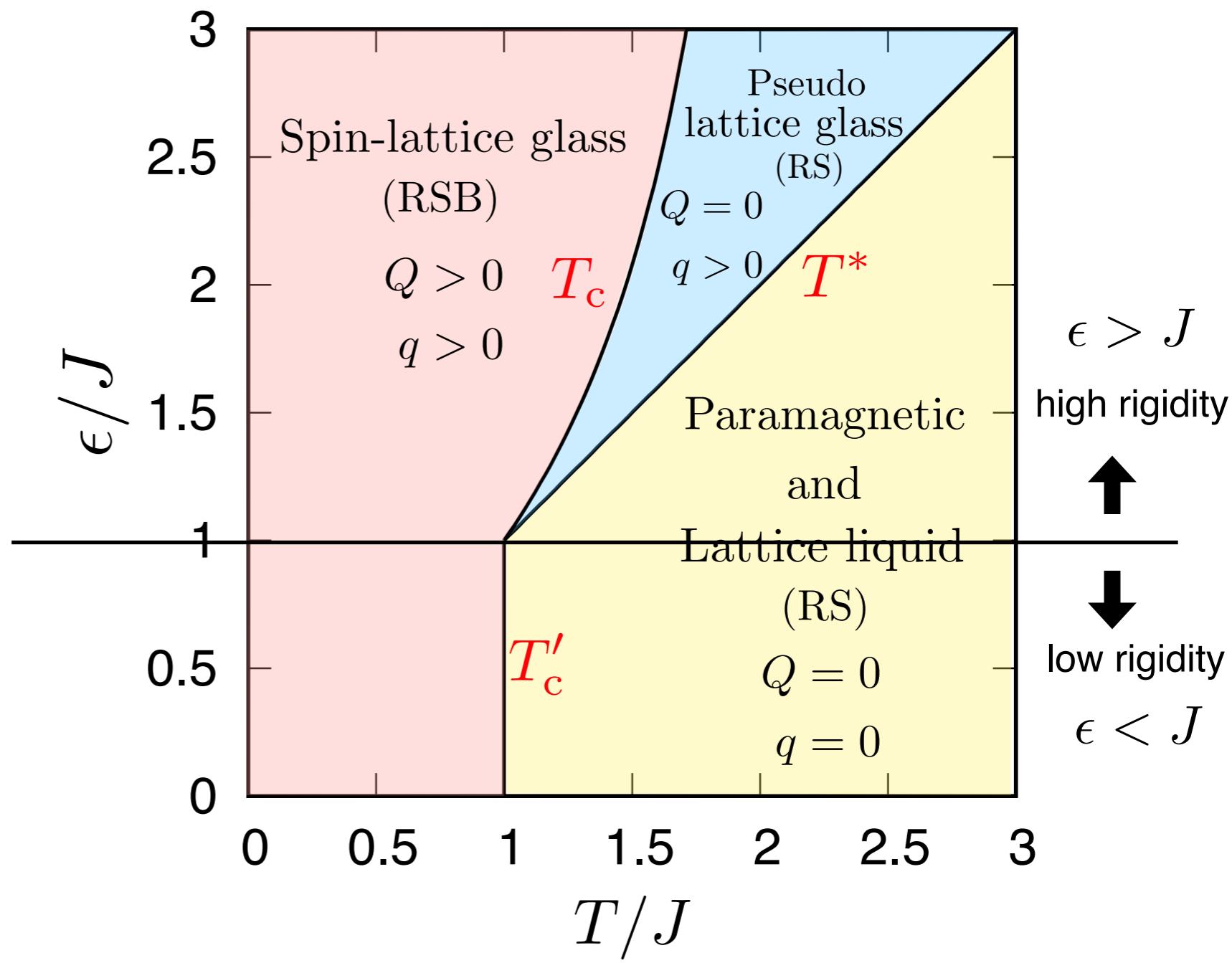
pseudo lattice glass



spin-lattice glass

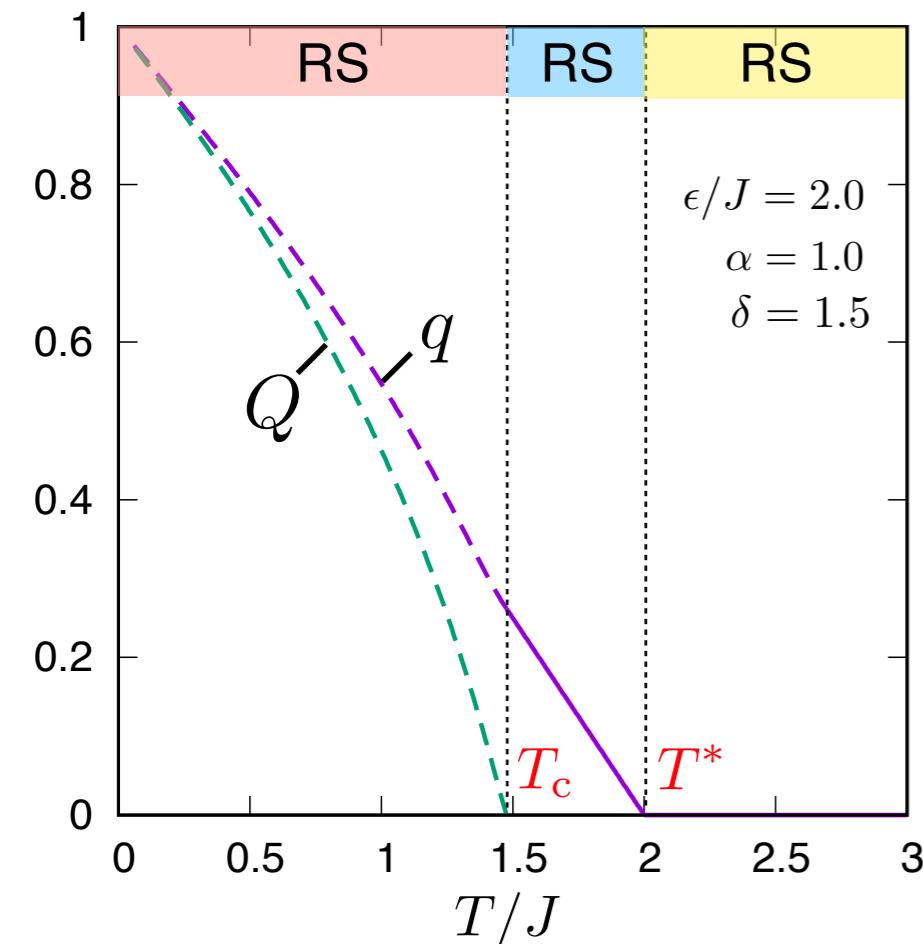


$\delta = 1.5$ (spin-lattice coupled)



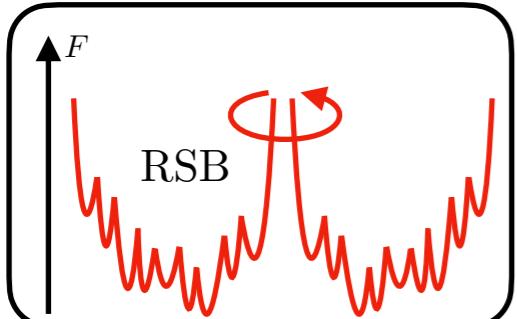
Order parameter (RS ansatz)

high rigidity

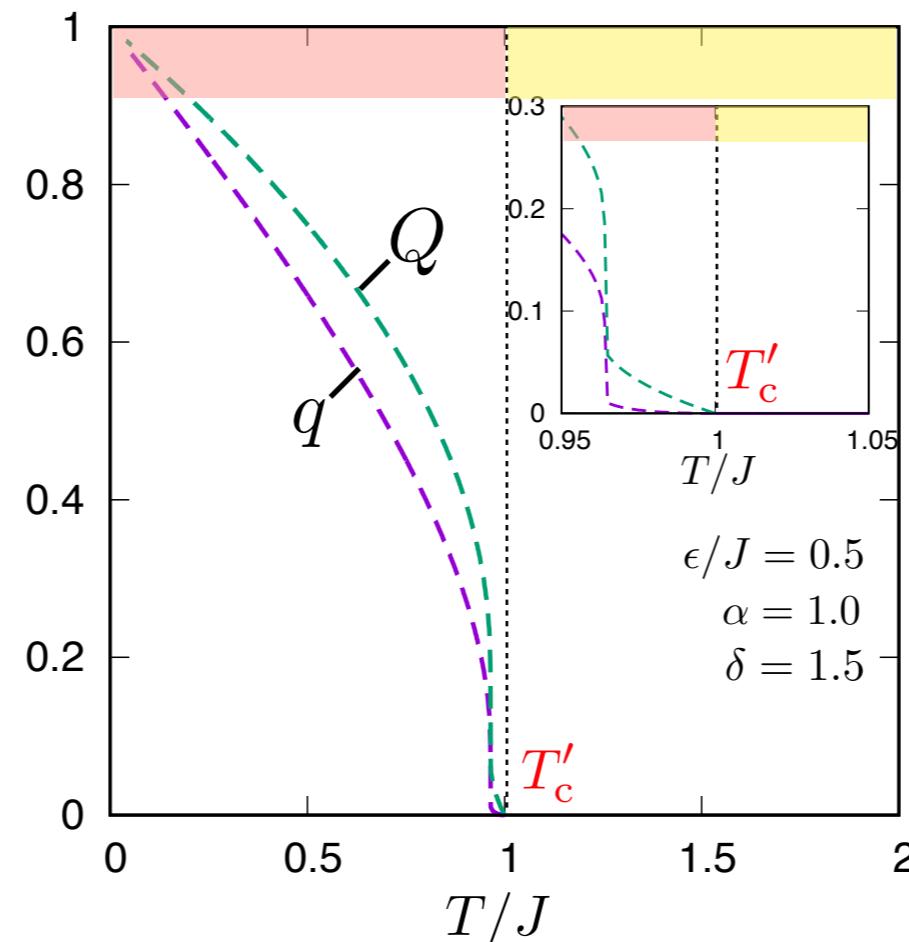


$$\lambda_{\min} = -2(\beta J \delta)^2 Q$$

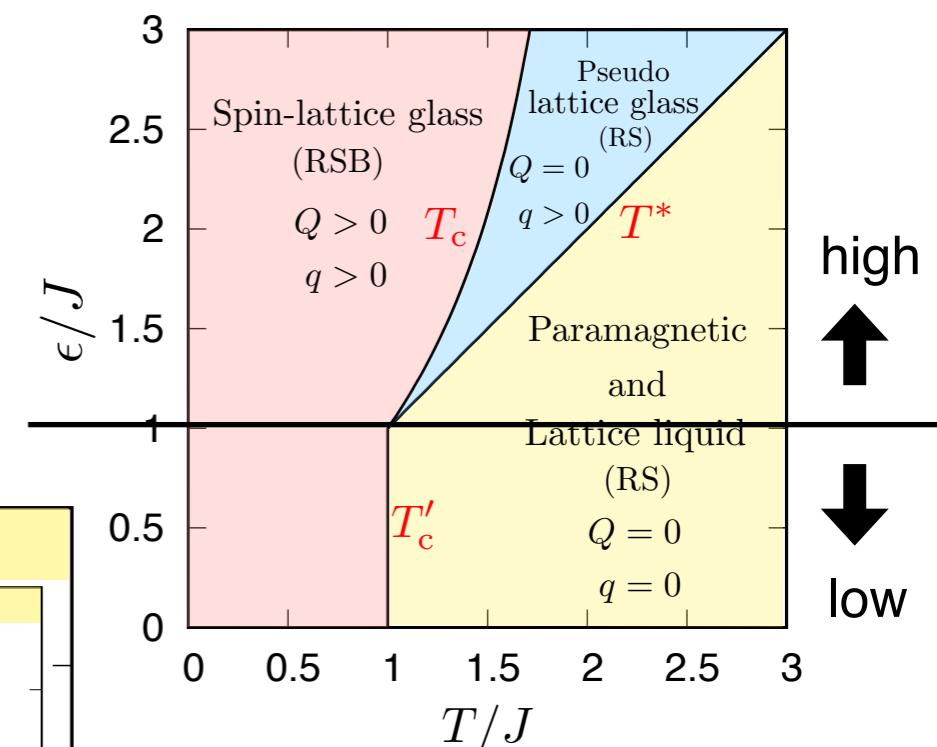
spin-lattice glass



low rigidity



$$\lambda_{\min} = -\frac{4(\beta J \delta)^4 Q^2}{1 - (\beta \epsilon)^2}$$



- Internal energy
- Heat capacity
- Glass susceptibility

Summary

We constructed a disorder-free spin-lattice glass model.

- Numerical simulation

=> Simultaneous spin-lattice glass transition

- divergence of relaxation times of spins and lattices at common T_c
- diverging features of nonlinear magnetic and dielectric susceptibilities around T_c

KM, C. Hotta and H. Yoshino Phys. Rev. Lett. **124**, 087201 (2020).

- Mean-field theory

=> Complex free-energy landscape due to spin-lattice coupling

- replica symmetry breaking (RSB) occurs in the spin-lattice glass phase

KM and H. Yoshino Phys. Rev. B **107**, 054412 (2023).

See also KM, C. Hotta and H. Yoshino Phys. Rev. Research **4**, 033157 (2022).