Thermodynamic signature of two-step melting of $\sqrt{3}\times\sqrt{3}$ order

Singular ferromagnetic suscepbility of the triangular lattice transverse-field Ising antiferromagnet

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Geometric frustration of exchange interactions

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- Triangles frustrate Néel order
- Geometry Scompetition between leading exchange interactions

Frustration spawns novel states

- Quenching of leading J is
 J cannot pick ground state at classical level
- Sub-dominant interactions & quantum fluctuations Solution
 Variety of novel low temperature states

Single ion anisotropy can be large

- Single ion anisotropy $-D(\mathbf{S} \cdot \mathbf{n})^2$ can dominate over J
- Pyrochlore spin ice Ho₂Ti₂O₇ (Ho³⁺, (L + S) = 8) Easy axes n point outward from center of each tetrahedron D ~ 50K, J ~ 1K Harris et. al., Phys. Rev. Lett. 79, 2554 (1997)
- Kagome Nd-langasite Nd₃Ga₅SiO₁₄ (Nd³⁺, (L + S) = 9/2)
 Easy axis perpendicular to lattice plane, J ~ 2K, D ~ 10K
 Robert *et. al.*, Physica B 2006
- $J \ll D$ is classical

study leading quantum effects in a J/D expansion

Anisotropy amplies frustration

Isotropic spins on a triangle



Easy-axis **n** and triangular motifs...



Wannier's triangular lattice model

• $H = J \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j - D \sum_i (S_i^z)^2$, with D >> J on the triangular lattice.

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- To leading order $S_i^z = \pm S \rightarrow \sigma = \pm 1$ $H \approx JS^2 \sum_{\langle ij \rangle} \sigma_i \sigma_j$
- Minimum energy configurations?

Minimally frustrated configurations



One frustrated bond per triangle

 Honeycomb lattice dimer model (one dimer touching each honeycomb site)

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Honeycomb lattice dimer model: One dimer touching each honeycomb vertex

Classic problem in graph-theory/combinatorics/statistical mechanics

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Ising 'liquid' in $T \rightarrow 0$ limit

Calculation of Stephenson (64) gives

$$\langle \sigma(\mathbf{r})\sigma(\mathbf{0})\rangle \sim \frac{A}{r^{9/2}} + \frac{B\cos\left(2\pi(x+y)/3\right)}{\sqrt{r}}$$

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- Spins neither freeze, nor fluctuate independently.
- Instead, form highly correlated "spin liquid".

Understanding this result:

- Dimers, heights, and Ising models of frustration
- (Obvious) connection to odd Ising gauge theories

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Connection to Kosterlitz-Thouless theory

Spins to dimers to electric fields



"Electric field" $\mathbf{e}_{A \rightarrow B} = n_{AB} - 1/3$ dimer constraint: Gauss law (!). Youngblood and Axe (1980)

From dimers to microscopic heights H(R)

$$\boldsymbol{e}_{l} = \boldsymbol{H}_{L(l)} - \boldsymbol{H}_{R(l)} \tag{1}$$



Henley 1990s

From microscopic H(R) to coarse-grained h(r)

- Coarse-grain: Average over local rearrangements
- ► Locality: What happens "outside" cannot affect what happens "inside". $h(r) \rightarrow h(r) + 1$ (Field theorists: "compactification radius")

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► Lattice translations and $2\pi/6$ rotation $h(r) \rightarrow h(r) + 1/3$, $h(r) \rightarrow -h(r)$

Ising spins in terms of h(r)

3*H* jumps by odd (even) number whenever one crosses an unfrustrated (frustrated) bond

 $\sigma(R) = \exp(-3\pi i H(R))$ (if $\sigma(R = 0) = +1$, and H(R = 0) = 0)



Dimer crossed \rightarrow spin unchanged; empty link \rightarrow spin flipped $\sigma(R) = \exp(i\pi \sum_{l} (1 - n_{l})) \sigma(0)$ $\sigma(R) = \exp(\frac{2\pi}{3}i(X + Y) - i\pi H(R))$ Fradkin et al (2004), KD (2009)

Effective action and operators

► Fewer flippable plaquettes \rightarrow larger "tilt" $S_{\text{eff}} = \frac{\pi}{g} (\nabla h)^2 + \lambda_6 \cos(6\pi h) + \dots$ Coarse-grained representation of spins: $\sigma(r) \sim Ae^{i\mathbf{Q}\cdot r}e^{-i\pi h(r)} + Be^{-3i\pi h(r)} + h.c.$ Three-sublattice order parameter $\psi \sim e^{i\pi h} \equiv e^{i\theta}(!)$

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T > 0: Odd Ising gauge theory and Kosterlitz-Thouless vortices

- Nonzero temperature: Fully frustrated triangle → three dimers touching honeycomb site.
- "Electric field *E*_{A→B} = *n*_{AB} 1/3 no longer divergence-free But violations are 0 mod 2
 Field-theory language: Configuration space of odd-Ising gauge theory
- ► Heights no longer single valued Three dimers touching honeycomb site \rightarrow vortex/antivortex in $\theta = \pi h$

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• T = 0: Vortex-free *xy* model for θ with 6-fold anisotropy T > 0: Vortices allowed

Picture for T = 0 power-law ordered phase

► In state with long-range three-sublattice order, θ feels $\lambda_6 \cos(6\theta)$ potential.

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Locks into values $2\pi m/6$ (resp. $(2m + 1)\pi/6$) in ferri (resp. antiferro) three-sublattice ordered state

In power-law three-sublattice ordered state λ₆ does not pin phase θ

 θ spread uniformly $(0, 2\pi)$

But vortices absent.

RG description

• Fixed point action: $S = \frac{1}{4\pi g} (\nabla \theta)^2$

► For $g > \frac{1}{9}$ $\lambda_6 \cos(6\theta)$ *irrelevant* along fixed line $\bowtie \langle \psi^*(r)\psi(0) \rangle \sim \frac{1}{r^{\eta(T)}}$ with $\eta = g$ Relies on absence of vortices at T = 0

Jose, Kadanoff, Kirkpatrick, Nelson (1977)

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Easy-axis antiferromagnets on triangular lattices



Natural tripartite structure

Perturbations/quantum fluctuations easily stabilize this order...

Three-sublattice order on the triangular lattice



 $\psi = |\psi| e^{i heta} = -\sum_{ec{R}} e^{i \mathbf{Q} \cdot ec{R}} S^z_{ec{R}}$

Ferri vs antiferro order distinguished by the choice of phase $\boldsymbol{\theta}$

Ferri: $\theta = 2\pi m/6$, Antiferro: $\theta = (2m + 1)\pi/6$ (m = 0, 1, 2...5)

Very recent: TmMgGaO₄ (Thulium [Xe] 4f¹³ 6s²) Tm³⁺ non-Kramers $J_z = \pm 6$

Princeton, Augsburg, Fudan (2017-18) *cf: talk by Liu (Gang Chen group) in this workshop Intrinsic transverse field* Γ_{int} : σ_{eff}^{χ} not a dipole

Prototypical example of order-by-(quantum) disorder

- *H*_{TFIM} = J ∑_{⟨ij⟩} σ^z_i σ^z_j − Γ ∑_i σ^x_i on the triangular lattice small Γ ISS Long-range order at three-sublattice wavevector Q
- ► Ordering of "antiferro" type → (+, -, 0) antiferro order provides maximum "room" for quantum fluctuations

Moessner, Sondhi, Chandra (2001), Isakov & Moessner (2003)

Another example: S = 1 with easy-axis single-ion anisotropy

- ► $H_{AF} = J \sum_{\langle ij \rangle} \vec{S}_i \cdot \vec{S}_j D \sum_i (S_i^z)^2$ on triangular lattice Closely related to effective model for AgNiO2 (Seabra & Shannon '11)
- ► Low-energy physics for $D \gg J$: $H_b = -\frac{J^2}{D} \sum_{\langle ij \rangle} (b_i^{\dagger} b_j + h.c.) + J \sum_{\langle ij \rangle} (n_i - \frac{1}{2})(n_j - \frac{1}{2}) + \dots$ KD & Senthil '06
- ► Low-temperature state for D ≫ J: "supersolid" state of hard-core bosons at half-filling. Auerbach & Murthy (97), Heidarian & KD, Melko, Wessel...(05)

 Implies: ferri three-sublattice order in S^z + "ferro-nematic" order in S²_⊥ (Simple easy-axis version of Chandra-Coleman (1991) "spin-nematic" ideas)

Symmetry breaking transitions: Generalities

- Symmetry-breaking state characterized by long-range correlations of "order-parameter" Ô
- phenomenological Landau free energy density \$\mathcal{F}[\heta]\$
 Expanding \$\mathcal{F}\$ in powers of \$\heta\$ (symmetry allowed terms)

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► Neglecting spatial variation & fluctuations: phase transition → change in minimum of *F*

Fluctuation effects at continuous transitions:

 More complete description of long-wavelength physics: Include (symmetry allowed) gradient terms in *F* Integrate over all possible order parameter configurations

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In most cases: Corrections to mean-field exponents

Symmetries are (usually) decisive:

Transformation properties of Ô determine nature of continuous transition

Landau-theory for melting of three-sublattice order

►
$$\mathcal{F} = K |\nabla \psi|^2 + r |\psi|^2 + u |\psi|^4 + \lambda_6 (\psi^6 + \psi^{*6}) + ...$$

Connection with six-state clock models
 $Z = \sum_{\{\rho_i\}} \exp[\sum_{\langle ij \rangle} V(\frac{2\pi}{6}(\rho_i - \rho_j))]$
Each $\rho_i = 0, 1, 2, ...5$
 $V(x) = K_1 \cos(x) + K_2 \cos(2x) + K_3 \cos(3x)$
Cardy (1980)

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Simplest lattice model

$$H_{\mathrm{xy}} = -J_{\mathrm{xy}} \sum_{\langle ec{r}ec{r}'
angle} \cos(heta_{ec{r}} - heta_{ec{r}'}) - h_6 \sum_{ec{r}} \cos(6 heta_{ec{r}}) \; .$$

(higher harmonics $J^{(p)}$ (p = 2, 3) left out of H_{xy} for simplicity)

Melting scenarios for three-sublattice order

- Analysis (Cardy 1980) of generalized six-state clock models

 Three generic possibilities of relevance here:
 Two-step melting, with power-law ordered intermediate phase OR
 - **3-state Potts transition** to ferromagnetic phase followed by loss of ferromagnetism via Ising transition at higher temperature.. or vice-versa...

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OR

First-order transition (always possible!)

Melting of three-sublattice order in various examples

- Antiferro three-sublattice order in triangular lattice transverse field Ising model
 Two-step melting (Isakov & Moessner '01)
- Ferrimagn. three-sublattice order in triangular lattice-gas models of monolayer films

Two-step melting

D.P. Landau '83

 Ferri. three-sublattice order in Kagome Ising antiferromagnets With second-neighbour ferro couplings: Two step melting Wolf & Schotte '88
 With long-range dipolar couplings: Three-state Potts transition

Moller & Moessner '09, Chern, Mellado, Tchernyshyov '11

Need scattering experiment to detect power-law version of Bragg peaks

Or

Real-space data by scanning some local probe + Lots of image-processing



Alternate thermodynamic signature(!)

Singular thermodynamic susceptibility to *uniform* easy-axis field
 B:

$$\chi_u(B) \sim \frac{1}{|B|^{\rho(T)}}$$

$$p(T) = \frac{4 - 18\eta(T)}{4 - 9\eta(T)} \text{ for } \eta(T) \in (\frac{1}{9}, \frac{2}{9})$$
So $p(T)$ varies from 2/3 to 0 as T increases from T_{c1} to just below T_{c2}

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(KD PRL '15)

Recall: picture for power-law ordered phase

In state with long-range three-sublattice order, θ feels $\lambda_6 \cos(6\theta)$ potential.

Locks into values $2\pi m/6$ (resp. $(2m + 1)\pi/6$) in ferri (resp. antiferro) three-sublattice ordered state for $T < T_{c1}$

- In power-law three-sublattice ordered state for T ∈ (T_{c1}, T_{c2}), λ₆ does not pin phase θ θ spread uniformly (0, 2π)
- But vortices continue to be irrelevant Distinction between ferri and antiferro three-sublattice order lost for $T \in (T_{c1}, T_{c2})$ Ferromagnetic response part of the time...

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Recall: More formally

Fixed point free-energy density: ^{*F*_{KT}}/_{*k*_B*T*} = ¹/_{4πg}(∇θ)² with g(T) ∈ (¹/₉, ¹/₄) corresponding to T ∈ (T₁, T₂)

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• $\lambda_6 \cos(6\theta)$ irrelevant along fixed line

•
$$\langle \psi^*(r)\psi(0)\rangle \sim \frac{1}{r^{\eta(T)}}$$

with $\eta(T) = g(T)$

Jose, Kadanoff, Kirkpatrick, Nelson (1977)

General argument—I

Starting point: Ferrimagnetic three-sublattice order also involves uniform magnetization m

More complete theory should treat m and ψ on equal footing

- Symmetries allow coupling term λ̃₃m(ψ³ + ψ∗³) augment F_{kT} with gapped free-energy density F_{ferro}(m): F_{ferro}(m) + λ₃m cos(3θ)
- λ_3 formally irrelevant along fixed line \mathcal{F}_{KT}

Physics of two-step melting unaffected—m "goes for a ride..."

But ...

 \rightarrow

General argument—II

• *m* "inherits" power-law correlations of $cos(3\theta)$: $C_m(r) = \langle m(r)m(0) \rangle \sim \frac{1}{r^{9\eta(T)}}$

•
$$\chi_L \sim \int^L d^2 r C_m(r)$$
 in a finite-size system at $B = 0$

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•
$$\chi_L = \chi_{\text{reg}} + bL^{2-9\eta(T)}$$
 for $\eta(T) \in (\frac{1}{9}, \frac{2}{9})$
Diverges with system size at $B = 0$

General argument—III

- Uniform field $B > 0 \rightarrow$ additional term $h_3 \cos(3\theta)$ in \mathcal{F}_{KT}
- ▶ Strongly relevant along fixed line, with RG eigenvalue 2 9g/2

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- Implies finite correlation length $\xi(B) \sim |B|^{-\frac{2}{4-9\eta}}$
- $\chi_u(B) \sim |B|^{-\frac{4-18\eta}{4-9\eta}}$ for $\eta(T) \in (\frac{1}{9}, \frac{2}{9})$

Test in prototypical example



In power-law ordered phase of $H_{\rm TFIM}$ on triangular lattice (Biswas & KD PRB '18)

Test in KT phase of easy-axis S = 1 triangular lattice AFM



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In power-law ordered phase of H_b (Heidarian & KD EPJB '18)

Also interesting:-Multicritical melting

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KT phase can
   Pinch-off at multicritical point \mathcal{M}_{?}, giving way to three-state Potts
   criticality. c_{M_2} = ?
   OR
   Pinch-off at multicritical point \mathcal{M}_{Clock}, giving way to first-order
   transition line.
   (KD PRL '15)
• \mathcal{M}_{\text{Clock}} previously known, not \mathcal{M}_{?}
   Note: Conjecture (Dorey-Tateo-Thompson '96) relates \mathcal{M}_{\text{Clock}} to
   self-dual Z_6 c = 1.25 CFT (Zamolodchikov-Fateev '85)
   \rightarrow c_{\mathcal{M}_{Clock}} = 1.25
   (KD PRL '15)
```

Also interesting: Multicritical melting

- How does the KT phase pinch-off for specific cases?
 - Evidence for *M*_{Clock} on the triangular lattice (Rakala, Shivam, Desai, & KD *in prep.*)
 - Similar results on Kagome lattice systems Conjecture for M_? in triangular bilayers (Rakala & KD unpublished)

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References:
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KD, PRL 115, 127204 (2015)

Some loose ends—I

Some loose ends...Three sublattice order and its melting in S=1 easy axis triangular antiferromagnet, and in classical Ising models on the triangular lattice

Is three-sublattice ordering of S^z in H_{AF} ferri or antiferro?

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 Natural expectation: Quantum fluctuations induce antiferro order (like in the transverse field Ising model)

Initial confusion: Ordering will be antiferro three-sublattice order *e. g.* Melko *et. al.* (2005)

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Actual state has ferrimagnetic three-sublattice order



Heidarian & KD PRL '05

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Early work: Triangular lattice-gas models for monolayer films on graphite

► Three-sublattice long-range order of noble-gas monolayers on graphite Birgeneau, Bretz, Chan, Vilches, Wiechert...(1970—1990) $H_{J_1J_2} = J \sum_{\langle ij \rangle} \sigma_i^z \sigma_j^z - J_1 \sum_{\langle \langle ij \rangle \rangle} \sigma_i^z \sigma_j^z - J_2 \cdots - B \sum_i \sigma_i^z$ Long-range three-sublattice ordering (wavevector **Q**) at low temperature D. P. Landau (1983)

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Test in J1-J2model



In power-law ordered phase of $H_{J_1J_2}$ ($R = -(J_1 + J_2)/J$ and $\kappa = (J_2 - J_1)/J$) (Geet Rakala & KD *in prep.*)

Some loose ends—II

Some loose ends...multicritical melting



Multicritical melting of three-sublattice order



More complete coarse-grained description

$$H_{\rm eff} = H_{\rm xy} + H_{\rm Ising} - J_{\theta\tau} \sum_{\vec{r}} \tau_{\vec{r}} \cos(3\theta_{\vec{r}}) ,$$

where $H_{\rm Ising} = -J_{\rm Ising} \sum_{\langle \vec{r}\vec{r}' \rangle} \tau_{\vec{r}} \tau_{\vec{r}'} - h \sum_{\vec{r}} \tau_{\vec{r}} ,$
 $H_{\rm xy} = -J_{\rm xy} \sum_{\langle \vec{r}\vec{r}' \rangle} \cos(\theta_{\vec{r}} - \theta_{\vec{r}'}) - h_6 \sum_{\vec{r}} \cos(6\theta_{\vec{r}}) ,$

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with $h \propto B$. (KD PRL '15)

The argument...

Start with known phase diagrams of H_{xy} and H_{Ising} and build in effects of J_{θτ}

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- When τ orders, H_{xy} sees effective three-fold symmetric perturbation h_{3eff} cos(3θ_r) with h_{3eff} ~ (τ)
- ► When $e^{i\theta}$ orders, H_{Ising} sees effective field $h_{\text{eff}}\tau_{\vec{r}}$ with $h_{\text{eff}} \sim \langle \cos(3\theta) \rangle$

The "new" multicritical point $M_{?}$

- c-theorem argument: $1 \le c \le \frac{3}{2}$
- To search:

 $J_{xy} = h_6 = 1.0, J_{\theta\tau} = 0.25$ Parametrize: $J_{\text{Ising}} = f_{xy} T_{\theta_1} / T_{\tau}$ and $T = f_I f_{xy} T_{\theta_1}$ [with $T_{\theta_1} = 1.04$ and $T_{\tau} = 3.6409$]

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Multicritical melting at $M_{?}$



$$\begin{split} & [f_{xy}^{\mathcal{M}_{7}}, f_{l}^{\mathcal{M}_{7}}] \approx [1.5570(8), 1.0061(5)] \\ & C_{2\theta} \left[C_{3\theta} \right] \text{ rescaled by a factor of 7 [factor of 10]} \\ & \eta_{3\theta} = \eta_{\tau} = 0.201(20), \, \eta_{\theta} = 0.258(5), \, \text{and} \, \eta_{2\theta} = 0.353(6). \\ & (\text{KD '15}) \end{split}$$

Speculation (aka wishful thinking?)

 If relative strength of first/second neighbour exchange tunable relative to long-range dipolar part in artificial kagome-ice:
 Could tune melting to multicritical point M₂...

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Computations challenging due to long-range interactions

$\mathcal{M}_?$ vs \mathcal{M}_{clock}

- Conjecture (Dorey '96): M_{clock} corresponds to c = 1.25 self-dual Z₆ CFT constructed by Zamolodchikov-Fateev ('85).
- Conjecture yields exponents at \mathcal{M}_{clock} : $\eta_{3\theta} = 3/8$, $\eta_{2\theta} = 1/3$, and $\eta_{\theta} = 5/24$. $\eta_{2\theta}$ and $\eta_{3\theta}$ very different from values at $\mathcal{M}_{?}$ Recall: at $\mathcal{M}_{?}$, $\eta_{3\theta} = \eta_{\tau} = 0.201(20)$, $\eta_{\theta} = 0.258(5)$, and $\eta_{2\theta} = 0.353(6)$.

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Test of conjectured exponents for \mathcal{M}_{clock}



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Results on Cardy's six-state clock model (Rakala, Shivam, & KD in prep.)

Schematic of pinch-off in triangular lattice Ising AFM



Evidence for \mathcal{M}_{clock} in triangular Ising AFM



R=2.0000

(Rakala, Shivam, & KD in prep.)

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Some loose ends—III

Some loose ends...previous work on Kagome systems



Three-sublattice order on the Kagome lattice



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Ising models for "Artificial Kagome-ice"

$$\blacktriangleright H_{\text{Kagome}} = J \sum_{\langle ij \rangle} \sigma_i^z \sigma_j^z - J_1 \sum_{\langle \langle ij \rangle \rangle} \sigma_i^z \sigma_j^z - J_2 \dots$$

- Only nearest-neighbour couplings → classical short-range spin liquid (Kano & Naya 1950)
- Second-neighbour ferromag. couplings destabilize spin liquid (Wolf & Schotte 88)
 Ferrimagnetic three-sublattice order at low *T*.
- "Artificial Kagome-ice: Moments M_i = σ_i^zn_i
 (n_i at different sites non-collinear)
 Expt: Tanaka *et. al.* (2006), Qi *et. al.* (2008), Ladak *et. al.* (2010,11)
 Theorem Mallan Massanan (2000). Charm et. al. (2011)

Theory: Moller, Moessner (2009), Chern et. al. (2011)

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