Melting of three-sublattice order

Kedar Damle, ISSP, UTokyo (April 2017) TIFR Mumbai



S. Biswas, D. Heidarian, G. Rakala (TIFR), S. Shivam

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Recap: Local moments in Motterials

► Band-theory of solids → electron-waves occupying modes determined by electrostatic potential of nuclear array

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- Strong e-e interactions \rightarrow Failure of band picture
- Electron particles localized on lattice sites charge frozen, spin remains dynamical

Recap: Antiferromagnetic exchange



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Recap: A Goodenough description

► Without spin-orbit: Isotropic exchange interactions. $E = J \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j$ J > 0When is J > 0, large?

Are nearest neighbour interactions dominant? Difficult (quantum chemistry/ab-initio studies) questions Thumb-rule answers: Goodenough-Kanamori-Anderson rules J.B. Goodenough, *Magnetism and the Chemical Bond (1963)*

Complications

- Spin-orbit coupling λ spin anisotropy terms
- Orbital degeneracy Interplay between orbital structure and spin physics *e.g.* Vanadium spinels (Tsunetsugu & Motome 2003)

Recap: Néel order

▶ Bipartite lattice and nearest neighbour J > 0Spins spontaneously pick axis **n** and $\langle \vec{S}_{\vec{r}} \rangle = (-1)^{\vec{r}}$ **n** Néel (antiferromagnetic) Order

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Geometric Frustration



- Triangles in nearest-neighbour connectivity *frustrate* Néel order
- Geometry induces *competition* between leading exchange interactions

Frustration spawns novel states

- Quenching of the leading exchange J
 J cannot pick ground state at classical level
- Sub-dominant interactions & quantum fluctuations play major role Opens the door for variety of novel low temperature states

Frustrated magnets: Plethora of lattices and materials

- ► Triangular lattice: S = 1 AgNiO₂ (Ni²⁺), S = 1/2 Cs₂CuCl₄ (Cu²⁺)...
- ► Kagome: S = 5/2 Fe jarosite (Fe³⁺), S = 1/2 Herbertsmithite ZnCu₃(OH)₆Cl₂ (Cu²⁺), S = 1 Ni₃V₂O₈ (Ni²⁺)...
- Pyrochlore, pyrochlore-slapb S = 3/2 SrCr_{9p}Ga_{12-9p}O₁₉ Cr³⁺ (SCGO)...

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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
- So it makes sense to study leading quantum effects in a J/D expansion
 Not our focus today

Anisotropy amplifies 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



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One frustrated bond per triangle



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

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

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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

From dimers to microscopic heights H(R)



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

 Locality: What happens "outside" cannot affect what happens "inside".

 $h(r) \rightarrow h(r) + 1$

More of a redundancy than a symmetry. (Field theorists: "compactification radius")

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- Translation symmetry: $h(r) \rightarrow h(r) + 1/3$
- Rotation by $2\pi/6$ about triangular site: $h(r) \rightarrow -h(r)$

Effective action for coarse-grained h(r)

► Fewer flippable plaquettes \rightarrow larger "tilt" $S_{\text{eff}} = \frac{\pi}{g} (\nabla h)^2 + \lambda_6 \cos(6\pi h) + \dots$

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Ising spins in terms of h(r)



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KT vortices and "odd Ising gauge theory"

- Nonzero temperature: Heights no longer single valued Vortex: h → h ± 2 ambiguity when three dimers touch honeycomb site (fully frustrated Ising triangle)
- Configuration space not dimer model, but model with odd number of dimers touching each 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

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Our focus: Easy-axis antiferromagnets on Kagome and triangular lattices



Natural tripartite structure

Three-sublattice order on the triangular lattice



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Three-sublattice order on the Kagome lattice



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On general symmetry grounds:

► Three ways in which this order can melt on heating Two-step melting (intermediate KT phase with power-law order for *T* ∈ (*T*_{c1}, *T*_{c2})) OR

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Three-state Potts transition followed by Ising transition OR

Single first-order transition (always possible!)

Our results—Landau-Ginzburg analysis:

- ► Thermodynamic signature of KT phase: $\chi_{\hat{n}}(B) \sim 1/|B|^{p(T)}$ with $p(T) \in (\frac{2}{3}, 0)$ for $T \in (T_{c1}, T_{c2})$.
- KT phase can

Pinch-off at multicritical point $M_?$, giving way to three-state Potts criticality. $c_{M_?} = ?$

OR

Pinch-off at multicritical point \mathcal{M}_{Clock} , giving way to first-order transition line.

• $\mathcal{M}_{\text{Clock}}$ previously known, not $\mathcal{M}_{?}$

Note: Conjecture (Dorey-Tateo-Thompson '96) relates M_{Clock} to self-dual $Z_6 c = 1.25 \text{ CFT}$ (Zamolodchikov-Fateev '85)

$$\rightarrow c_{\mathcal{M}_{\text{Clock}}} = 1.25$$

Our results—Computations for microscopic models:

- Existence of KT phase in S = 1 triangular lattice antiferromagnets with moderate easy-axis (single-ion) anisotropy
- Quantitative verification of predicted singular susceptibility in KT phase in several cases
- How does the KT phase pinch-off for specific cases?
 - Preliminary evidence for M_{Clock} on the triangular lattice Similar, more preliminary results on Kagome lattice systems

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► Conjecture for *M*_? in triangular bilayers

Incipient three-sublattice order in triangular Ising AFM

► Recall: Power-law correlator in $T \to 0$ limit: $\langle \sigma_r^z \sigma_0^z \rangle \sim \frac{\cos(\mathbf{Q} \cdot \mathbf{r})}{r^{1/2}}$ Incipient order at three-sublattice wavevector $\mathbf{Q} = (2\pi/3, 2\pi/3)$ Stephenson (1964)

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Perturbations can stabilize this order...

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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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 $\mathbf{M}_i = \sigma_i^z \mathbf{n}_i$ (\mathbf{n}_i at different sites non-collinear) Expt: Tanaka *et. al.* (2006), Qi *et. al.* (2008), Ladak *et. al.* (2010,11) Theorem Maller Massesser (2000). Chara et. al. (2011)

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

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Prototypical example of order-by-(quantum) disorder

- ► $H_{\text{TFIM}} = J \sum_{\langle ij \rangle} \sigma_i^z \sigma_j^z \Gamma \sum_i \sigma_i^x$ on the triangular lattice 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)

Prospects for experimental realization slim? (large moments, dipolar couplings...)

S = 1 triangular lattice antiferromagnets with single-ion anisotropy (more promising)

- $H_{\rm AF} = J \sum_{\langle ij \rangle} \vec{S}_i \cdot \vec{S}_j D \sum_i (S_i^z)^2$ on triangular lattice
- ▶ 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: Three-sublattice order in S^z + "ferro-nematic" order in \vec{S}_1^2

(Simple easy-axis version of Chandra-Coleman (1991)

"spin-nematic" ideas)

(also related to Tsunetsugu-Arikawa (2006) proposal for $NiGa_2S_4)$

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 (2005)

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 \hat{O} determine nature of continuous transition

Landau-theory for melting of three-sublattice order

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

Connection with six-state clock models
$$Z = \sum_{\{p_i\}} \exp[\sum_{\langle ij \rangle} V(\frac{2\pi}{6}(p_i - p_j))]$$

Each $p_i = 0, 1, 2, \dots 5$
$$V(x) = K_1 \cos(x) + K_2 \cos(2x) + K_3 \cos(3x)$$

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 (2001)
- 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) Nature of melting transition in $S = 1 H_{AF}$?

 Prediction of Boninsegni & Prokofiev (2005)
 Three-state Potts transition
 Prediction based on argument about relative energies of different kinds of domain walls
 hard to get right at quantitative level

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Our answer from large-scale QMC simulations



Heidarian & KD (submitted to PRB)

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

Or

 $\label{eq:real-space} \begin{array}{l} \textit{Real-space} \mbox{ data by scanning some local probe} + \textit{Lots of image-processing} \end{array}$



Alternate thermodynamic signature(!)

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

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

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

KD (PRL 2015)

Recall: picture for power-law ordered phase

In state with long-range three-sublattice order, θ feels λ₆ cos(6θ) potential.
 Locks into values 2πm/6 (resp. (2m + 1)π/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 ∈ (T_{c1}, T_{c2})

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More formal RG description

► Fixed point free-energy density: $\frac{\mathcal{F}_{\text{KT}}}{k_B T} = \frac{1}{4\pi g} (\nabla \theta)^2$ with $g(T) \in (\frac{1}{9}, \frac{1}{4})$ corresponding to $T \in (T_1, T_2)$

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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 ...

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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

- 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_{TFIM} on triangular lattice Biswas, KD (*submitted to PRB*)

More complete coarse-grained description

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

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

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with $h \propto B$. KD (PRL 2015)

Phase diagram of $H_{\rm eff}$



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The argument...

Start with known phase diagrams of H_{xy} and H_{Ising} and build in effects of $J_{\theta\tau}$

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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 $\mathcal{M}_{?}$

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

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

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Multicritical melting at $M_{?}$



$$\begin{split} & [f_{xy}^{\mathcal{M}_{?}}, f_{I}^{\mathcal{M}_{?}}] \approx [1.5570(8), 1.0061(5)] \\ & C_{2\theta} \left[C_{3\theta}\right] \text{rescaled by a factor of 7 [factor of 10]} \\ & \eta_{3\theta} = \eta_{\tau} = 0.201(20), \, \eta_{\theta} = 0.258(5), \, \text{and} \, \eta_{2\theta} = 0.353(6). \\ & \text{KD (PRL 2015)} \end{split}$$

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 *M*₂...

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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}



Preliminary results on Cardy's six-state clock model (S. Shivam)

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Schematic of pinch-off in triangular lattice Ising AFM



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



R=2.0000

Preliminary results (Geet Rakala)

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Ongoing computations for six-state clock models: Sounak Shivam (IITB \rightarrow EPFL Lausanne)

+ Closely related multicritical point: Master's thesis of Nisheeta Desai (BITS-Goa \rightarrow Kentucky)

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Computational resources at TIFR