# Order by disorder and phase transitions

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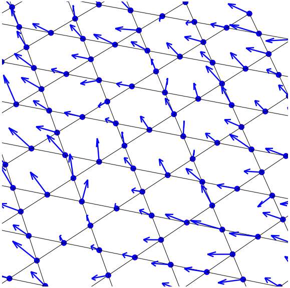
# Introduction - Phase transitions and frustrated classical spin systems

In the limit of large electron repulsion $U$ and integer filling, the **Hubbard model** has a Mott insulating phase, described by a **Heisenberg Hamiltonian**:

$$H = \sum_{\langle ij \rangle} J_{ij} \, \mathbf{S}_i \cdot \mathbf{S}_j .$$



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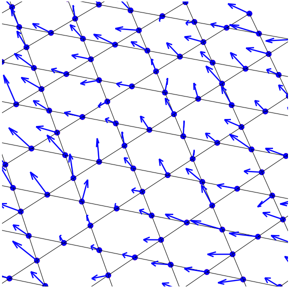

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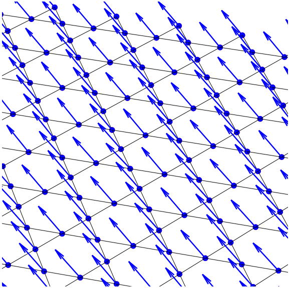

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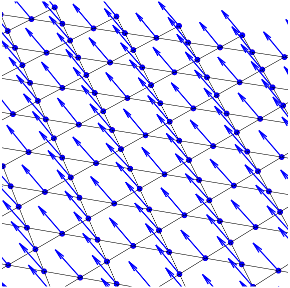

Such systems can develop a **long range order** at low $T$, breaking for example the spins rotational symmetry.

Bad example: ferromagnetic order in 2d.

**Mermin Wagner theorem:** a continuous symmetry cannot be broken at $T \neq 0$ in $d < 3$.

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**Mermin Wagner theorem:**
a continuous symmetry cannot be broken at $T \neq 0$ in $d < 3$.

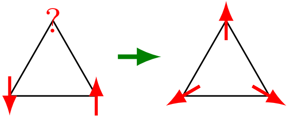
However, **low dimensions** host interesting phases.

Does it mean that no phase transition exists in 2d ?

$\rightarrow$ possibility to break **discrete symmetries** (2d Ising transition).
... possible with continuous spins ?

$\rightarrow$ need for complicated ground state !
... but how to find such ground states ?

**Frustration:**




In classical and quantum magnets, frustration leads to exotic ground states. It arises from the geometry of the lattice or of competing interactions.

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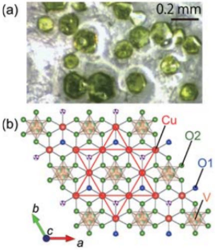
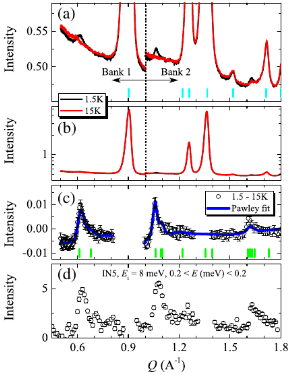
# Vesignieite $BaCu_3V_2O_8(OH)_2$

- Vesignieite, layers of $S = 1/2$ Cu atoms on a kagome lattice.
- Curie Weiss temperature $\simeq -77K$
  $\Longrightarrow$ possible $1^{st}$ neighbor AF !




Okamoto et al, JPSJ **78**, (2009)

Okamoto et al, PRB **83**, (2011)

Colman et al, PRB. **83**, (2011)

Quilliam et al, PRB. **84**, (2011)

Yoshida et al, J. Mater. Chem. **22**, (2012)

Yoshida et al, Chem. Mater. **29**, (2017)

- But uncompatible magnetic correlations (neutron scattering results).
  Magnetic peaks at 0.64, 1.08, and 1.65 $\AA^{-1}$, i.e. $\mathbf{k} = (\frac{1}{2}, 0, 0)$.
  Boldrin et al, PRL **121**, (2018)

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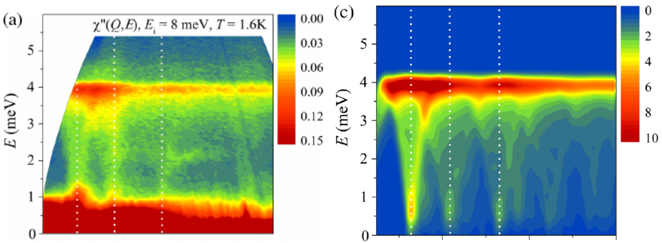
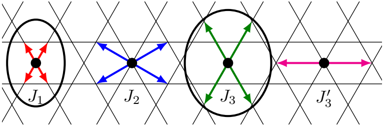



Boldrin et al, PRL **121**, 107203 (2018)




Spin wave calculations and chemical considerations
$\Longrightarrow$ dominant $J_3$ antiferromagnetic interactions ($> 15|J_1|$).

$$H = J_1 \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J_3 \sum_{\langle ij \rangle_3} \mathbf{S}_i \cdot \mathbf{S}_j.$$

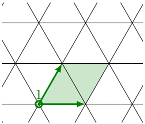
# The Luttinger-Tisza method

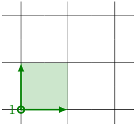
Translational invariance of the Hamiltonian $\longrightarrow$ Fourier transformation, with $\mathbf{r}$, $\mathbf{r}+\mathbf{v}$ from a Bravais lattice, $i$ and $j$ sites in each unit-cell.

$$H = \sum_{\mathbf{r}} \sum_{\mathbf{v},i,j} J_{i,j,\mathbf{v}} \mathbf{S}_{i,\mathbf{r}} \cdot \mathbf{S}_{j,\mathbf{r}+\mathbf{v}} = \sum_{\mathbf{q}} \sum_{i,j} \tilde{\mathbf{S}}_{i,\mathbf{q}} \tilde{J}_{i,j,\mathbf{q}} \tilde{\mathbf{S}}_{j,-\mathbf{q}}$$

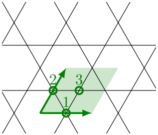
$$\mathbf{S}_{i,\mathbf{r}} = \frac{1}{\sqrt{N}} \sum_{\mathbf{q}} \tilde{\mathbf{S}}_{i,\mathbf{q}} e^{i\mathbf{q}\cdot\mathbf{r}}, \qquad \tilde{J}_{i,j,\mathbf{q}} = \sum_{\mathbf{v}} J_{i,j,\mathbf{v}} e^{i\mathbf{q}\cdot\mathbf{v}}$$

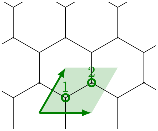
For a **Bravais lattice**, $\tilde{J}_{\mathbf{q}}$ is a scalar, and a GS is obtained from the lowest energy $\mathbf{q}$ mode $\longrightarrow$ spiral ground state.



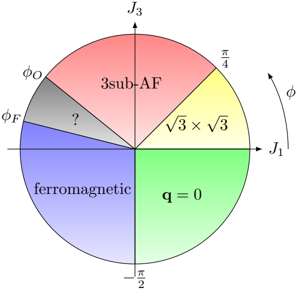



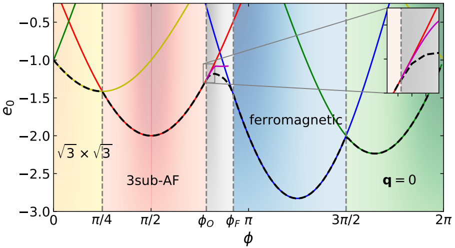

For **non-Bravais lattice**, $|\mathbf{S}_i|$ can be $\neq 1$.
Eigenvalues of $\tilde{J}_{\mathbf{q}}$ matrices give a **lower bound** for $E_{GS}$.



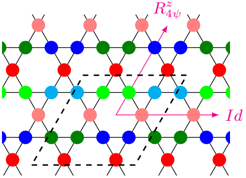



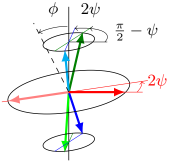
# Classical phase diagram at $T = 0$ (Heisenberg spins)

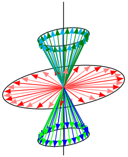



$\phi_O = \pi - \arctan \frac{1+\sqrt{5}}{4}$ separates the 3subAF phase from an *alternating conic state* (magenta energy), similar to Sklan et al, PRB **88**, 024407 (2013)



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# Accidental and non accidental degeneracies

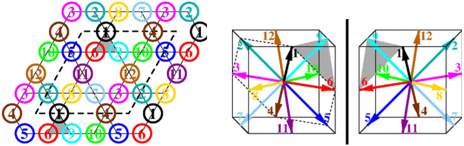
**Non accidental degeneracy**: all ground states (GSs) are **equivalent** up to a Hamiltonian symmetry (global spin rotation, lattice transformation, time reversal...).

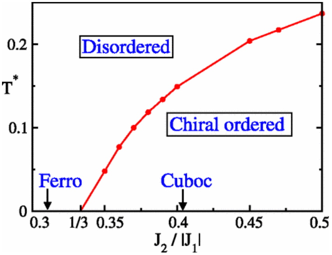
Mermin Wagner theorem: GSs related by a continuous symmetry are indistinguishable at $T \to 0^+$ in $d \geq 2$.

Thus, **phases transitions** only occur (in $d \geq 2$) when the set of GSs has several **connected components**

Example:

$$H = \sum_{\langle ij \rangle} \underbrace{J_1}_{<0} \mathbf{S}_i \cdot \mathbf{S}_j \sum_{\langle\langle ij \rangle\rangle} + \underbrace{J_2}_{>0} \mathbf{S}_i \cdot \mathbf{S}_j$$



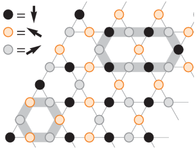



$SO(3) \times \mathbb{Z}_2 \longrightarrow$ chiral phase transition.

# Accidental and non accidental degeneracies

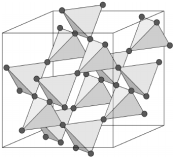
**Accidental degeneracy**: the GSs are not related by a Hamiltonian symmetry.

### Kagome $J_1$



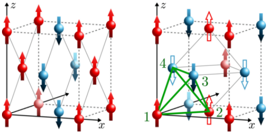

Cépas et al, PRB **84** (2011)

### Pyrochlore



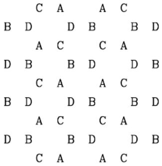
Moessner & Chalker, PRL **80** (1998)

### FCC $J_1$




Schick et al, PRB **106** (2022)

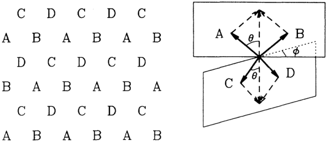
### Honeycomb $J_1 - J_2 - J_3$




Fouet et al, EPJB **20** (2001)

### Triangular $J_1 - J_2$

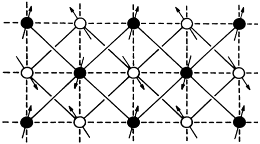
$J_1/8 < J_2 < J_1$




Lecheminant et al, PRB **52** (1995)

### Square $J_1 - J_2$

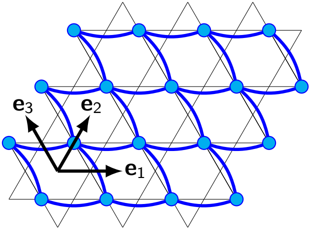
$J_1/2 < J_2$



Henley, PRL **77** (1989)

# The 3subAF phase

For $J_3 > 0$ and small $J_1$, same ground state as for $J_1 = 0$:
3 decoupled square lattices.



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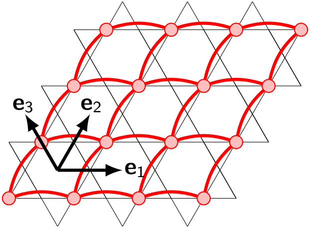

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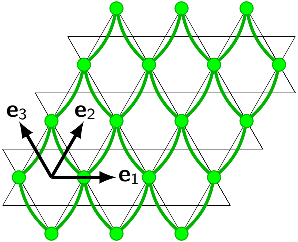

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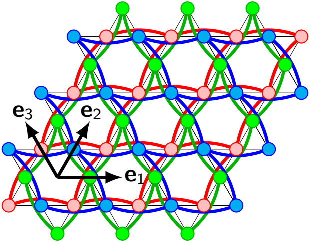

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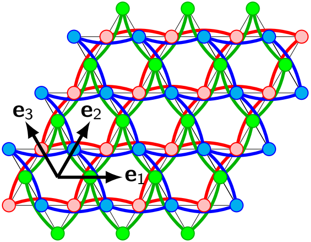


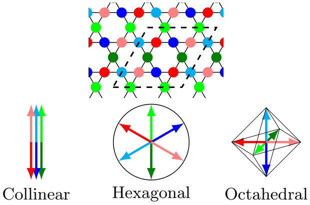

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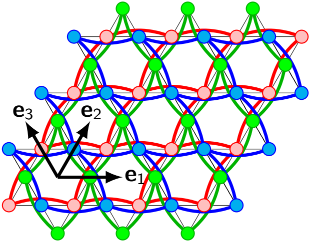
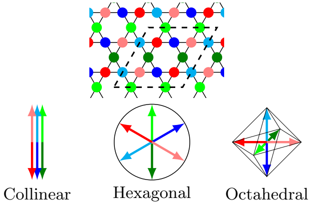
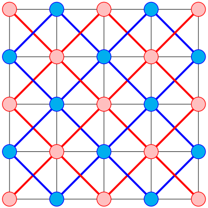
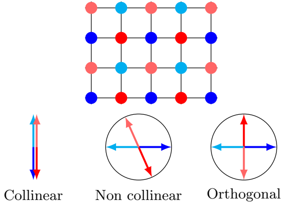



Same phenomena as on the square $J_1 - J_2$ lattice for small $J_1$:
2 decoupled square lattices.





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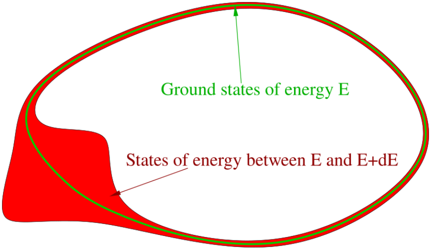
# The order by disorder phenomena

In case of accidental degeneracy, un-equivalent states have the same ground state energy, but are not related by a symmetry.

Then, fluctuations around them are different, and have different energies.

$\longrightarrow$ states with the largest number of low energy excitations are selected for $T \to 0^+$.



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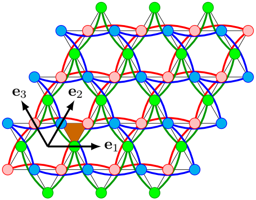


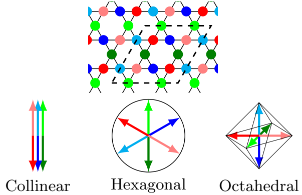


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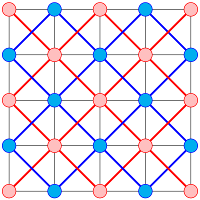

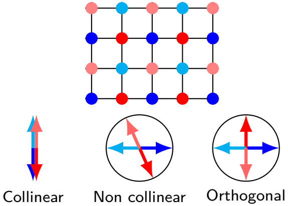

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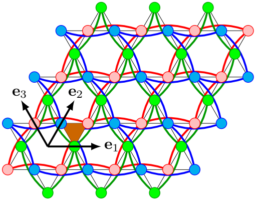


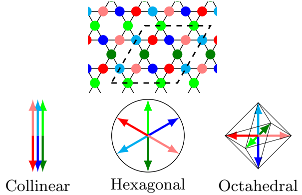


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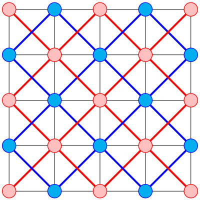

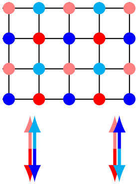

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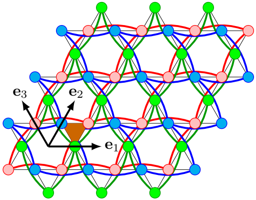


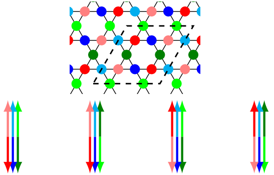

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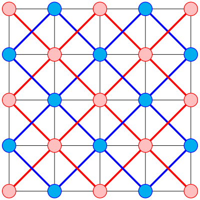

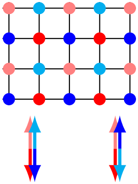

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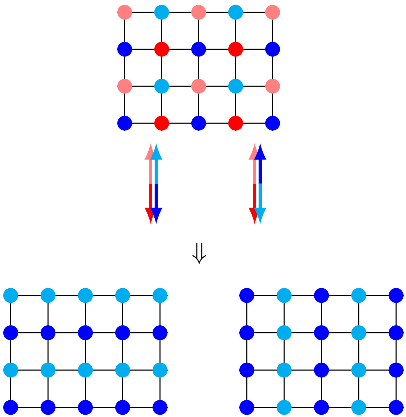
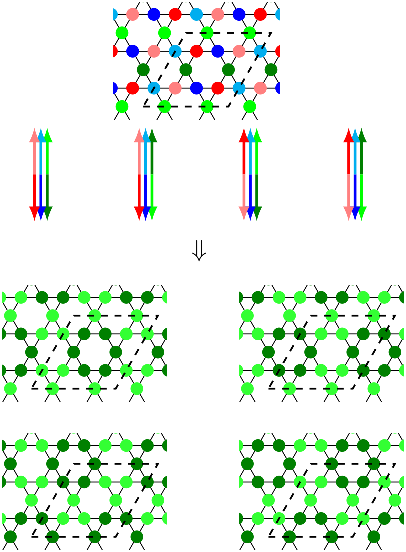
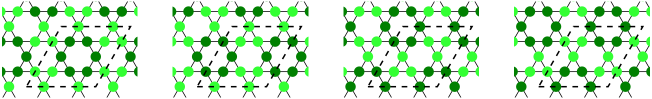




Square: Ising parameter,

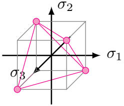
Kagome: $q = 4$ Potts parameter.

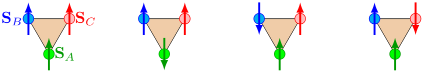
# The $q = 4$ Potts order parameter



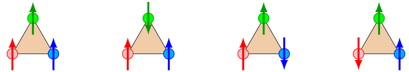
The translational discrete symmetry is broken $\rightarrow$ need for an order parameter.

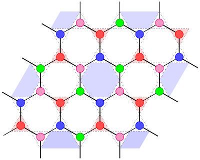
$$\sigma = \begin{pmatrix} \sigma_1 = \mathbf{S}_B \cdot \mathbf{S}_C \\ \sigma_2 = \mathbf{S}_C \cdot \mathbf{S}_A \\ \sigma_3 = \mathbf{S}_A \cdot \mathbf{S}_B \end{pmatrix}$$

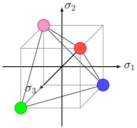



$$\begin{pmatrix} +1 \\ +1 \\ +1 \end{pmatrix} \quad \begin{pmatrix} +1 \\ -1 \\ -1 \end{pmatrix} \quad \begin{pmatrix} -1 \\ +1 \\ -1 \end{pmatrix} \quad \begin{pmatrix} -1 \\ -1 \\ +1 \end{pmatrix}$$

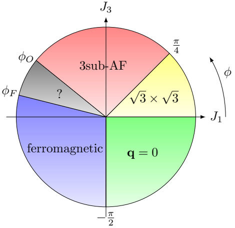


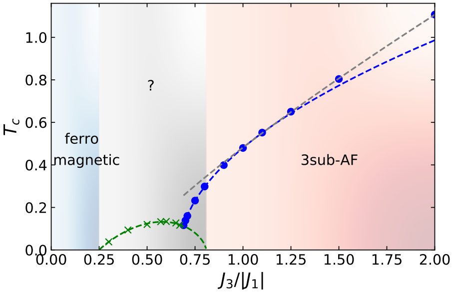


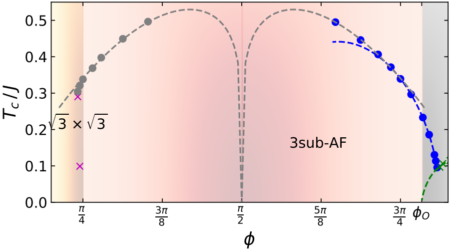



- Definition of an order parameter $\sigma$ on each triangle, that is alternated in the collinear ground state $\Longrightarrow \Sigma$.

# Classical phase diagram from MC simulations



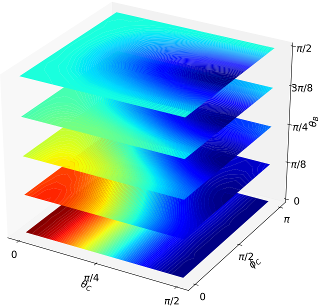

- Parallel tempering + reweighting + finite size scaling
- $K_4$ phase transition in the 3subAF phase,
- $K_4$ finite-$T$ order in the grey and $\sqrt{3}\times\sqrt{3}$ phases.
- high $J_3$: $T_c \propto T$ (grey dashed), small $J_3$: $T_c \propto \sqrt{J_3 - J_3^c}$ (blue dashed).

# Order by disorder: linear spin wave approximation

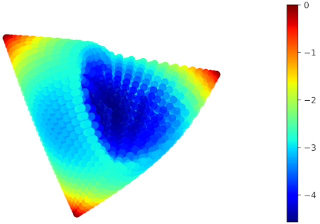
A classical order can be defined by 3 angles, with the spin directions on the 3 sublattices:

$$\mathbf{S}_A^0 = \pm \begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix}, \; \mathbf{S}_B^0 = \pm \begin{pmatrix} \sin\theta_B \\ 0 \\ \cos\theta_B \end{pmatrix}, \; \mathbf{S}_C^0 = \pm \begin{pmatrix} \sin\theta_C \cos\phi_C \\ \sin\theta_C \sin\phi_C \\ \cos\theta_C \end{pmatrix}.$$

Linear spin wave theory evidences thermal (through entropy) or quantum (through energy) selection of collinear ground states (favored states in red):




$\Delta S$ in the $(\theta_B, \theta_C, \phi)$ space,




in the $(\mathbf{S}_B^0\mathbf{S}_C^0,\ \mathbf{S}_C^0\mathbf{S}_A^0,\ \mathbf{S}_A^0\mathbf{S}_B^0)$ space

# PLAN

- Introduction - Phase transitions and frustrated spin systems

- The classical kagome $J_1 - J_3$ model and its phase diagram

- The 3sub-AF phase: an example of accidental degeneracy

- Thermal order by disorder and phase transitions

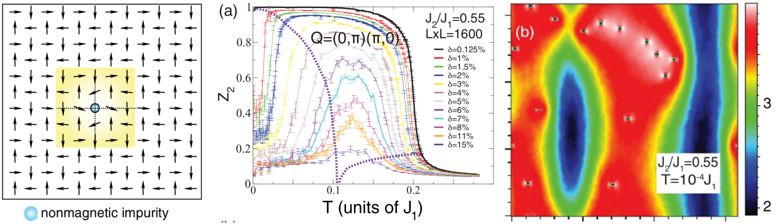
- Quench disorder and its effect on the phase diagram

# Effect of vacancies

Entropic effects tend to align collinearly spins, while magnetic vacancies act similarly to a local field that the AF square sublattices perpendicularly to it:

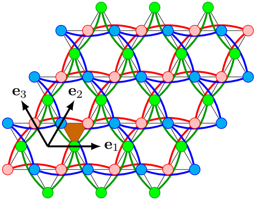
$$V_{ent} \propto -T \sum_{\langle i,j \rangle} (\mathbf{S}_i \cdot \mathbf{S}_j)^2, \qquad V_{imp} \propto \sum_{\langle i,j \rangle} (\mathbf{S}_i \cdot \mathbf{S}_j)^2.$$

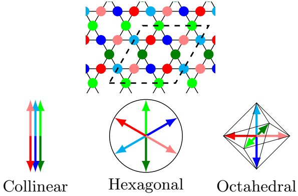
$J_1 - J_2$ **model on the square lattice** $\longrightarrow$ anticollinear state.



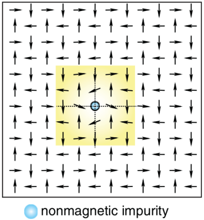

Weber and Mila, PRB *86* 184432 (2012)

# Effect of impurities in the 3subAF phase on the ground state (GS)

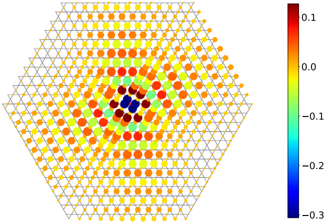



Least collinear GS $\longrightarrow$ octahedral order with 3 perpendicular sublattices.

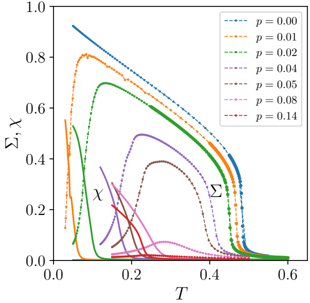


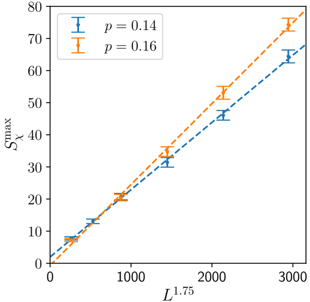

No broken $\mathbb{Z}_2$ symmetry




Chirality: a broken $\mathbb{Z}_2$ symmetry.

# The chiral phase

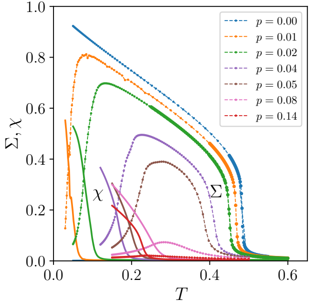


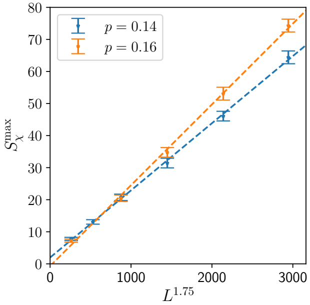




Simulations for $L = 96$.

Bottom: maximum of the susceptibilty $\mathcal{S}_\chi^{\max}$ versus $L^{1.75}$ (Ising universality class).

# The chiral phase

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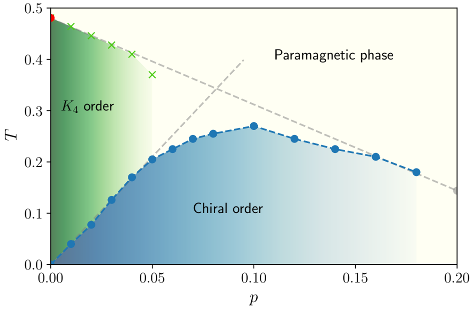
Presence of a clear chiral phase:



# The former $K_4$ phase

Two main categories of couples disorder/order:

Disorder in Quantum Many-Body Systems, Vojta, Ann. Rev. of Cond. Matt. Phys. *10* 1 (2019)

- **random-field type disorder**, which breaks the symmetry between ordered stated,

- **random-$T_c$ type disorder**, which preserves it, but induces spatial fluctuations in $T_c$.

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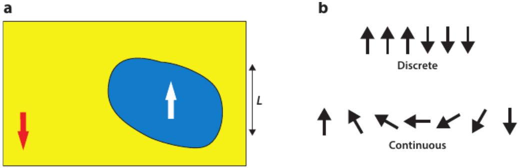
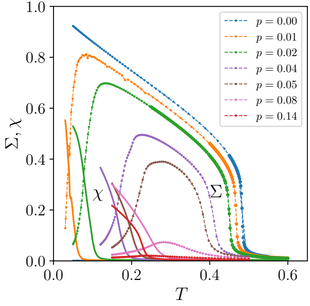
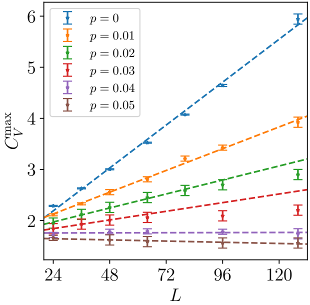
- **random-field type disorder**, which breaks the symmetry between ordered stated,
  Example: Ising model with random field
  Effect: kills the transition in $d \leq 2$ for a discrete order, $d \leq 4$ for a continuous order, by the Imry-Ma argument,



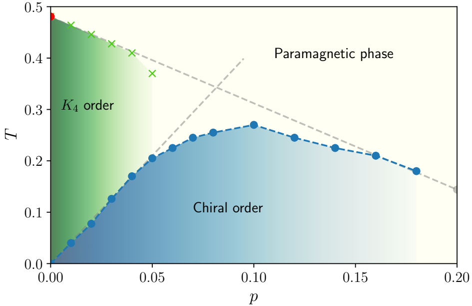

- **random-$T_c$ type disorder**, which preserves it, but induces spatial fluctuations in $T_c$.
  Example: Ising model with vacancies or random interactions
  Effect: softens the transition (no more first order transitions)

# The $K_4$ phase




No more $K_4$ phase transition, but a crossover for $p > 0$.



# Perspectives and conclusion

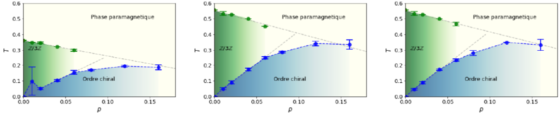
- We have identified a phase transition with $K_4$ order parameter in a $J_1 - J_3$ model, due to the order by disorder mechanism.

  Grison, Viot, Bernu, LM, PRB **102**, 214424 (2020)

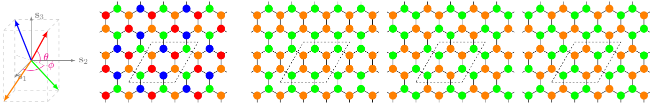
- Impurities destroy this order, but induce a chiral phase in the Ising universality class.

  Letouzé, Viot, LM, PRL **135**, 186504 (2025)

- This phenomena is also present on several other lattices.




(a) Réseau triangulaire (tétraédrique) (b) Réseau hexagonal (cubique) (c) Réseau hexagonal (tétraédrique)




Florian Couriol, LM, in preparation

Thank you for your attention

Collaborators (LPTMC): Florian Couriol, Coraline Letouzé, Vincent Grison, Bernard Bernu, Pascal Viot.