INSTITUT NANOSCIENCES ET CRYOGÉNIE

la recherche, ressource fondamentale

research - a fundamental resource

SBT | SCIB | SPINTEC | SPRAM | SPSMS | SP2M

inac.cea.fr

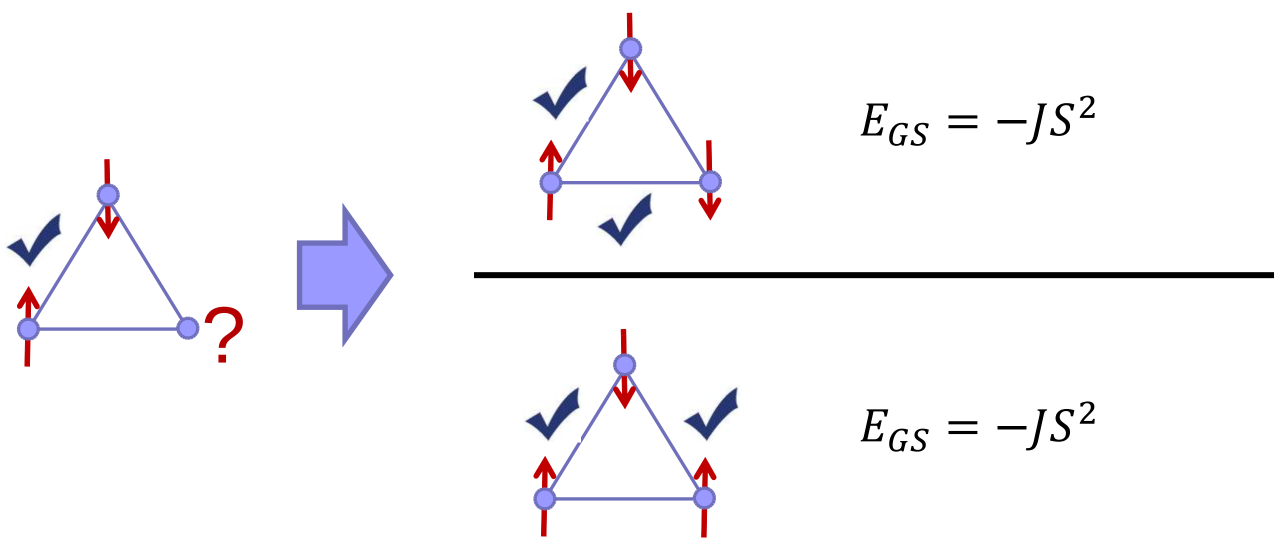
Order by structural disorder in Heisenberg triangular antiferromagnet

and other frustrated models

V. Maryasin, M. Zhitomirsky

Magnetic frustration

Competing interactions → Frustration → Ground state degeneracy



$$E_{GS} = -JS^2$$

$$E_{GS} = -JS^2$$

Outline

Pure Heisenberg TAFM

- Mean-field results
- Thermal ObD
- Phase diagram

HTAFM with impurities

- Model and setup
- Order by structural disorder

Numerical methods and results

- Ground state minimization
- Monte Carlo simulations
- Phase diagram

Beyond isotropic classical model

- Quantum ObD
- Easy-plane TAFM
- $Er_2Ti_2O_7$

INSTITUT NANOSCIENCES
ET CRYOGÉNIE

Outline

Pure Heisenberg TAFM

- Mean-field results
- Thermal ObD
- Phase diagram

HTAFM with impurities

Numerical methods and results

Beyond isotropic classical model

Heisenberg Triangular Antiferromagnet

TAFM: Kawamura, Miyashita, JPSJ (1984)
XY TAFM: Lee et al., PRB (1986)

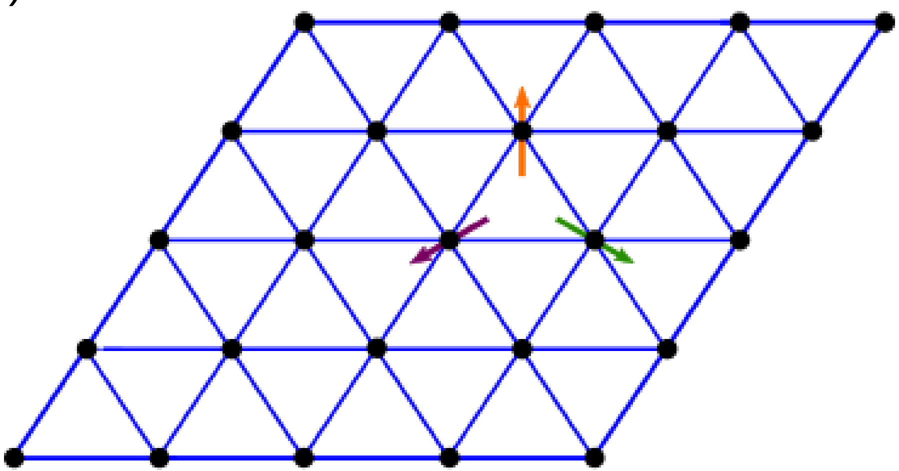
$$\hat{H} = J \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j$$

Condition for a GS

$$\mathbf{S}_\Delta = 0$$

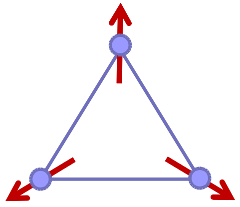
Local field, acting on each spin

$$H_{loc} = -JS \sum_{i=1}^{6} \cos\theta_{ij} = 3JS$$

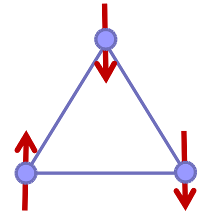


3 – sublattice $\sqrt{3} \times \sqrt{3}$ ordering

120° - configuration



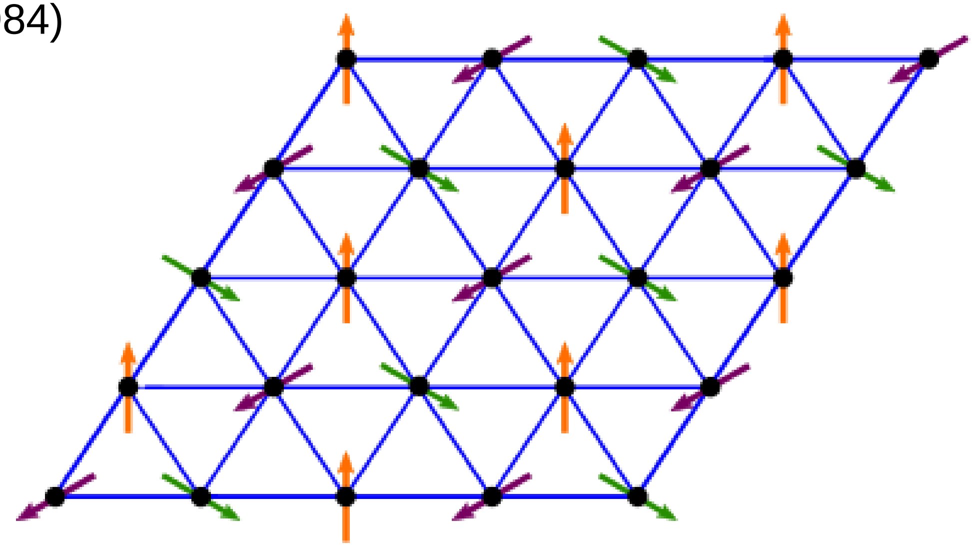
$$E_{GS} = -\frac{3}{2} JS^2$$



$$E_{GS} = -JS^2$$

Heisenberg Triangular Antiferromagnet

TAFM: Kawamura, Miyashita, JPSJ (1984)
XY TAFM: Lee et al., PRB (1986)



$$\hat{H} = J \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j$$

Condition for a GS

$$\mathbf{S}_\Delta = 0$$

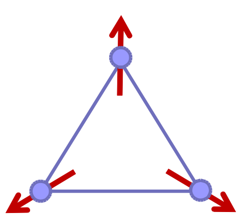
Local field, acting on each spin

$$H_{loc} = -JS \sum_{i=1}^{6} \cos\theta_{ij} = 3JS$$

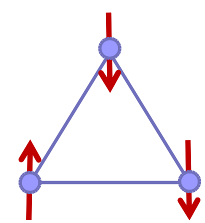
$NaCrO_2$, Cs_2CuBr_4

$RbFe(MoO_4)_2$, $Ba_3CoSb_2O_9$

120° - configuration



$$E_{GS} = -\frac{3}{2}JS^2$$



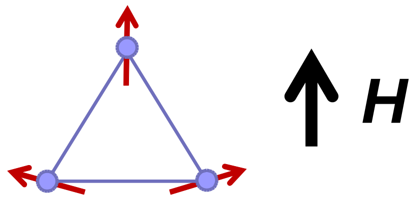
$$E_{GS} = -JS^2$$

Frustration in HTAFM

$$\hat{H} = J\sum_{\langle i,j\rangle} \mathbf{S}_i \cdot \mathbf{S}_j - \mathbf{H}\sum \mathbf{S}_i$$

Condition for a GS

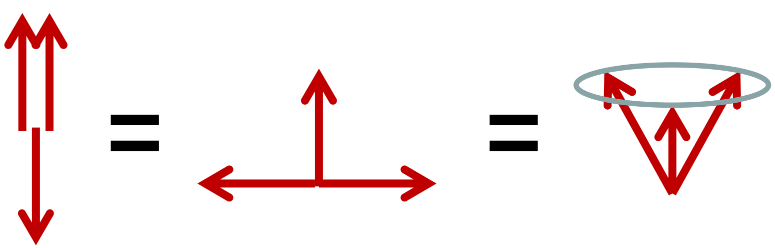
$$\mathbf{S}_\Delta = \frac{\mathbf{H}}{3J}$$



Local field, acting on each spin

$$H_{loc} = -JS\sum_{i=1}^{6} \cos\theta_{ij} + H\cos\theta_j = 3JS$$

Example: $H = 3JS$;

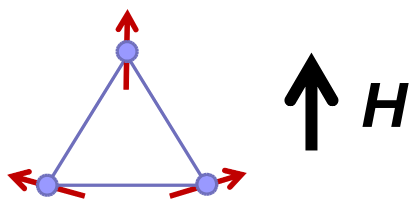


Frustration in HTAFM

$$\hat{H} = J \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j - \mathbf{H} \sum \mathbf{S}_i$$

Condition for a GS

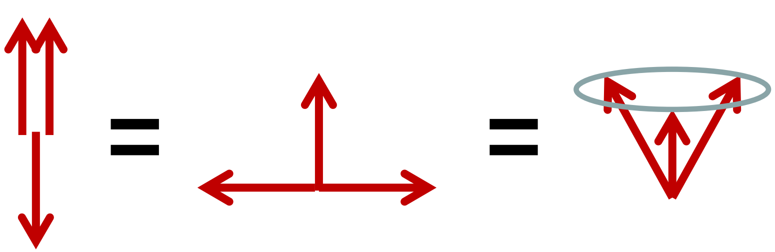
$$\mathbf{S}_\Delta = \frac{\mathbf{H}}{3J}$$



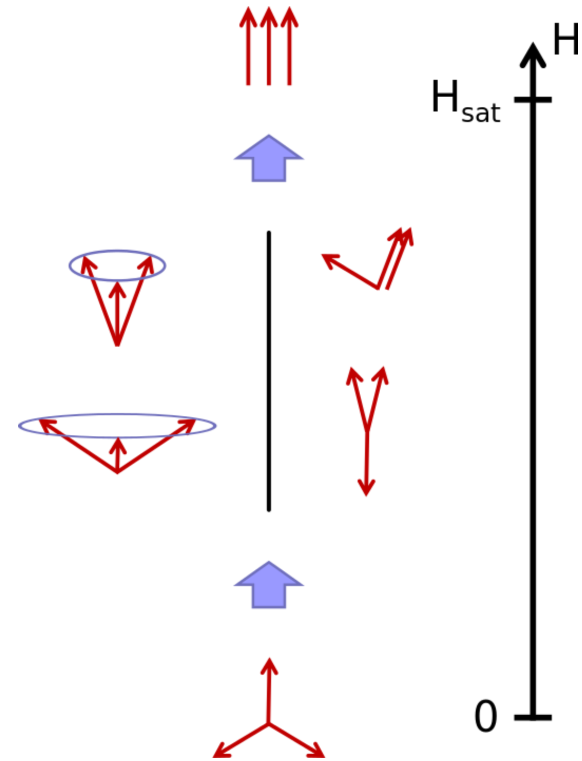
Local field, acting on each spin

$$H_{loc} = -JS \sum_{i=1}^{6} \cos\theta_{ij} + H\cos\theta_j = 3JS$$

Example: $H = 3JS$;



Accidental degeneracy: arbitrary orientation of spin plane with respect to $\mathbf{H}$



Order by Disorder

J. Villain *et al.*, J. de Physique, 1980
E. Shender JETP, 1982

Energetically degenerate ground states have different entropy or zero-point fluctuation spectrum

- Thermal order by disorder

$$F = E_{GS} - TS$$

- Quantum order by disorder

$$E_Q = E_{GS} + \frac{\hbar}{2}\sum_k \omega_k$$

Real space perturbation theory

Thermal order by disorder for a classical model

$$\hat{H} = J \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j - \mathbf{H} \sum \mathbf{S}_i$$

$$\hat{H} = \sum_{\langle i,j \rangle} J_{ij} \left[S_i{}^y S_j{}^y + \cos\theta_{ij} \left(S_i{}^z S_j{}^z + S_i{}^x S_j{}^x \right) + \sin\theta_{ij} \left(S_i{}^z S_j{}^x - S_i{}^x S_j{}^z \right) \right] - \mathbf{H} \sum_i \mathbf{S}_i$$

Angles θ_{ij} parameterize the ground state

Real space perturbation theory

Thermal order by disorder for a classical model

$$\hat{H} = J\sum_{\langle i,j\rangle} \mathbf{S}_i\cdot\mathbf{S}_j - \mathbf{H}\sum \mathbf{S}_i$$

$$\hat{H} = \sum_{\langle i,j\rangle} J_{ij}\left[S_i{}^y S_j{}^y + \cos\theta_{ij}\left(S_i{}^z S_j{}^z + S_i{}^x S_j{}^x\right) + \sin\theta_{ij}\left(S_i{}^z S_j{}^x - S_i{}^x S_j{}^z\right)\right] - \mathbf{H}\sum_i \mathbf{S}_i$$

$$\hat{H}_0 = -H_{\rm loc}\sum_i S_i{}^z \simeq \frac{1}{2}H_{\rm loc}\sum_i\left[S_i{}^{x2} + S_i{}^{y2}\right] \qquad \hat{V}_1 = J\sum_{\langle i,j\rangle}\left[S_i{}^y S_j{}^y + \cos\theta_{ij} S_i{}^x S_j{}^x\right]$$

Second order perturbation to F due to $\hat{V}_1$

$$F = F_0 - \frac{\langle V^2\rangle_0}{2T} + \cdots = F_0 - \frac{J^2 T}{2H_{\rm loc}{}^2}\sum_{\langle i,j\rangle}\left(1+\cos^2\theta_{ij}\right)$$

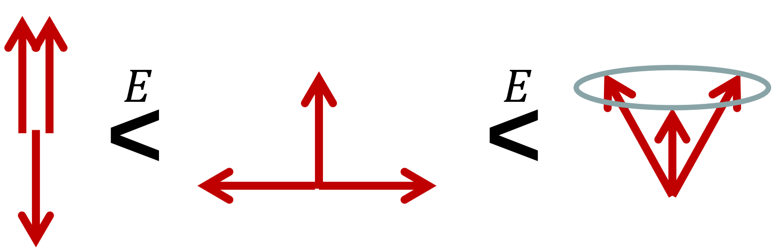
$$\Delta F = -\frac{T}{18}\sum_{\langle i,j\rangle}\left(\boldsymbol{S}_i\cdot\boldsymbol{S}_j\right)^2$$

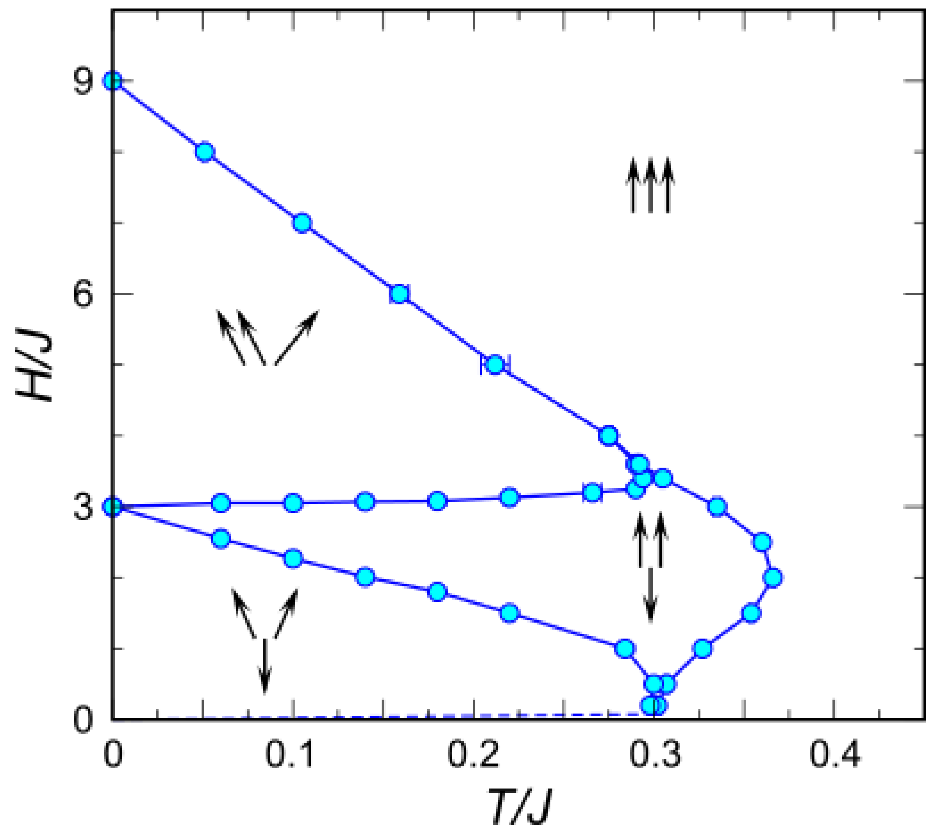
Order by disorder in classical HTAFM

Thermal fluctuations can be described by an effective biquadratic exchange term

$$\Delta F = -\frac{T}{18}\sum_{\langle i,j\rangle}\left(\boldsymbol{S}_i\cdot\boldsymbol{S}_j\right)^2$$

Thermal order by disorder selects the most collinear phases





Gvozdikova *et al.*, JPCM (2011)

Outline

Pure Heisenberg TAFM

HTAFM with impurities

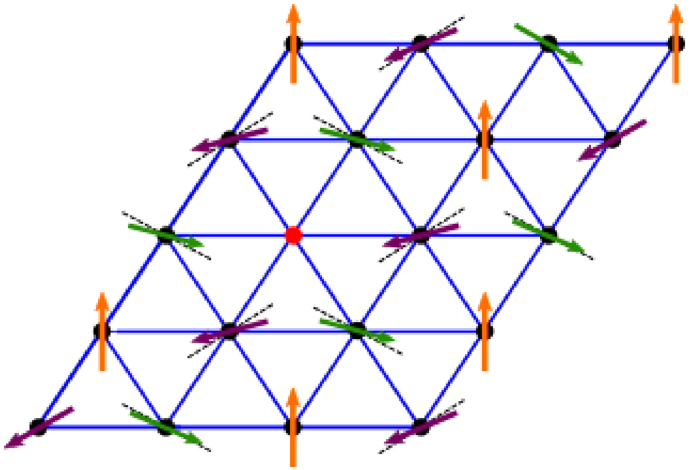
- Model and setup
- Order by structural disorder

Numerical methods and results

Beyond isotropic classical model

Structural disorder in frustrated magnets

- Nonmagnetic impurities (vacancies)
- Bond disorder



Weak bond disorder

$$J \to J_{ij} = J + \delta J_{ij}$$

$$\langle \delta J_{ij} \rangle = 0; \quad \langle \delta {J_{ij}}^2 \rangle = \delta J^2$$

δJ_{ij} - random and uncorrelated

Weak nonmagnetic impurities

$$S_i = 1 - \varepsilon; \quad p_i = 1$$

$$S_i = 1; \quad p_i = 0$$

$$\mathbf{S}_i \to \mathbf{S}_i \,(1 - p_i) + \mathbf{S}_i \, p_i (1 - \varepsilon)$$

$$J S_i S_j \to J S_i S_j \left[1 - \varepsilon\left(p_i + p_j\right) + \varepsilon^2 p_i p_j\right]$$

$$J_{ij} = J\left[1 - \varepsilon\left(p_i + p_j\right)\right]$$

INSTITUT NANOSCIENCES ET CRYOGÉNIE

Positive biquadratic coupling

Order by structural disorder for a classical model

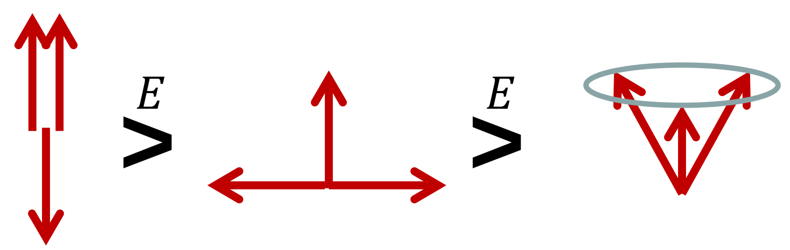
$$\hat{H} = \sum_{\langle i,j \rangle} (J + \delta J_{ij}) \left[S_i{}^y S_j{}^y + \cos\theta_{ij} \left(S_i{}^z S_j{}^z + S_i{}^x S_j{}^x \right) + \sin\theta_{ij} \left(S_i{}^z S_j{}^x - S_i{}^x S_j{}^z \right) \right] - \mathbf{H} \cdot \sum_i \mathbf{S}_i$$

$$\hat{H}_0 = \frac{1}{2} H_{\mathrm{loc}} \sum_i \left[S_i{}^{x2} + S_i{}^{y2} \right] \qquad \widehat{V_2} = \sum_{\langle i,j \rangle} \delta J_{ij} \sin\theta_{ij} \left(S_j{}^x - S_i{}^x \right)$$

Minimize the Hamiltonian over the deviations $S_i{}^x$

$$\Delta E = -\frac{\delta J^2}{2H_{\mathrm{loc}}} \sum_{\langle i,j \rangle} \sin^2\theta_{ij}$$

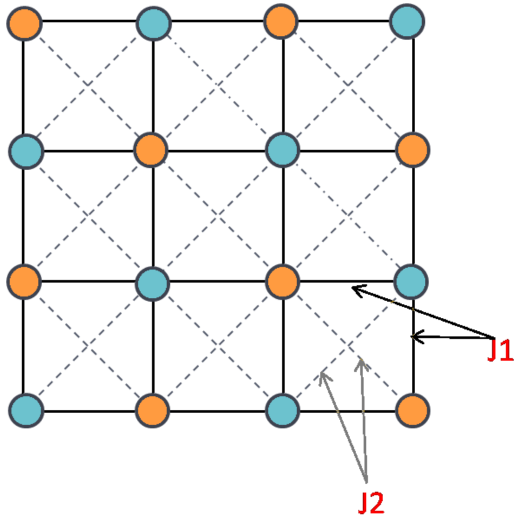
$$\Delta E = \frac{\delta J^2}{6J} \sum_{\langle i,j \rangle} \left(\boldsymbol{S_i} \cdot \boldsymbol{S_j} \right)^2$$



Order by structural disorder in $J_1 - J_2$ AFM

$J_1 - J_2$ model with $J_2 > 0.5\, J_1$

$$\hat{H} = J_1 \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J_2 \sum_{\langle\langle i,j \rangle\rangle} \mathbf{S}_i \cdot \mathbf{S}_j$$



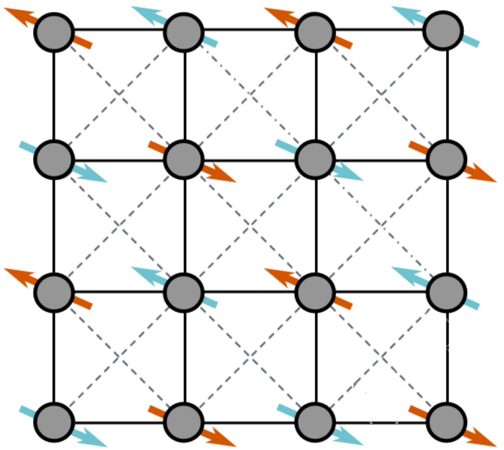
INSTITUT NANOSCIENCES
ET CRYOGÉNIE

Order by structural disorder in $J_1 - J_2$ AFM

$J_1 - J_2$ model with $J_2 > 0.5\, J_1$

$$\hat{H} = J_1 \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J_2 \sum_{\langle\langle i,j \rangle\rangle} \mathbf{S}_i \cdot \mathbf{S}_j$$

Collinear $\mathbf{Q} = (\pi, 0)$ or $\mathbf{Q} = (0, \pi)$ ordering – conventional ObD.

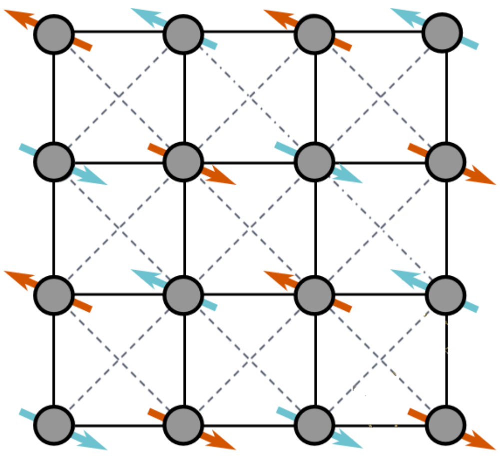


Order by structural disorder in $J_1 - J_2$ AFM

$J_1 - J_2$ model with $J_2 > 0.5\, J_1$

$$\hat{H} = J_1 \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J_2 \sum_{\langle\langle i,j \rangle\rangle} \mathbf{S}_i \cdot \mathbf{S}_j$$

Collinear $\mathbf{Q} = (\pi, 0)$ or $\mathbf{Q} = (0, \pi)$ ordering – conventional ObD.



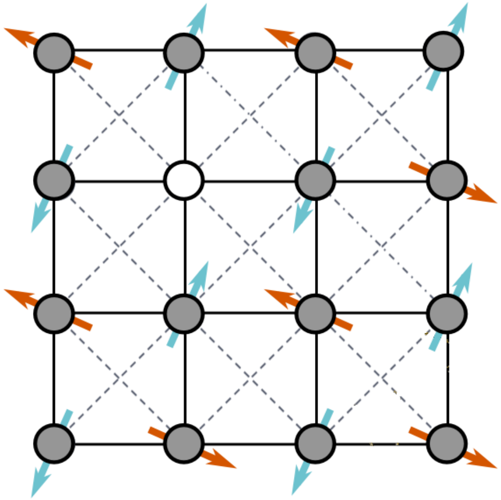
Nonmagnetic impurities in $J_1 - J_2$ model

C. Henley PRL (1989)

C. Weber, F. Mila PRB (2012)

Vacancy induces nonzero H_{loc} on a neighboring lattice

Anticollinear $\mathbf{Q} = (\pi, \pi)$ ordering –
a response of a Neel AFM to an external field.



Outline

Pure Heisenberg TAFM

HTAFM with impurities

Numerical methods and results

- Ground state minimization
- Monte Carlo simulations
- Phase diagram

Beyond isotropic classical model

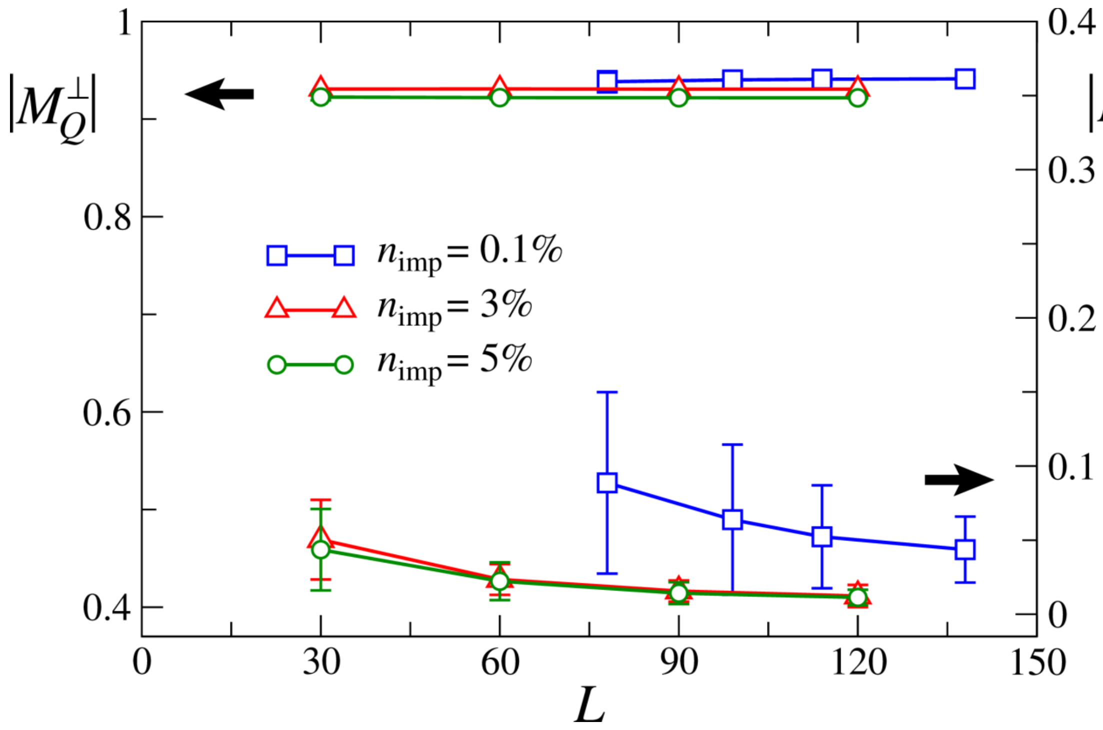
Ground state minimization

Periodic $L \times L$ rhombohedral cluster with $30 \leq L \leq 150$

Search for a global minimum of energy for a random impurity configuration

All measured quantities are averaged over 100 random impurity configurations

Staggered magnetization $\mathbf{M}_Q = \frac{1}{N}\sum_i \mathbf{S}_i e^{-i\mathbf{Q}r_i}$ $Q = \left(\frac{4\pi}{3}, 0\right)$

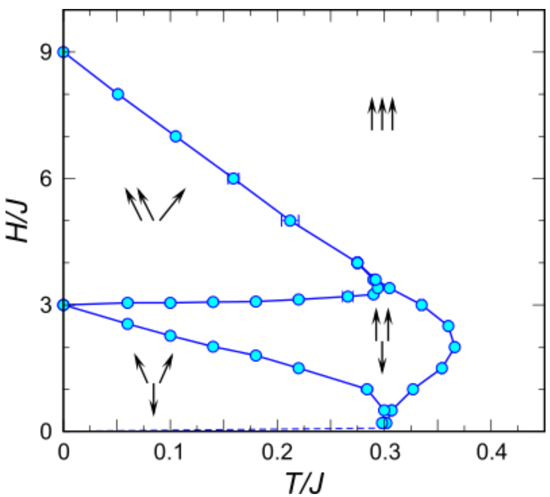


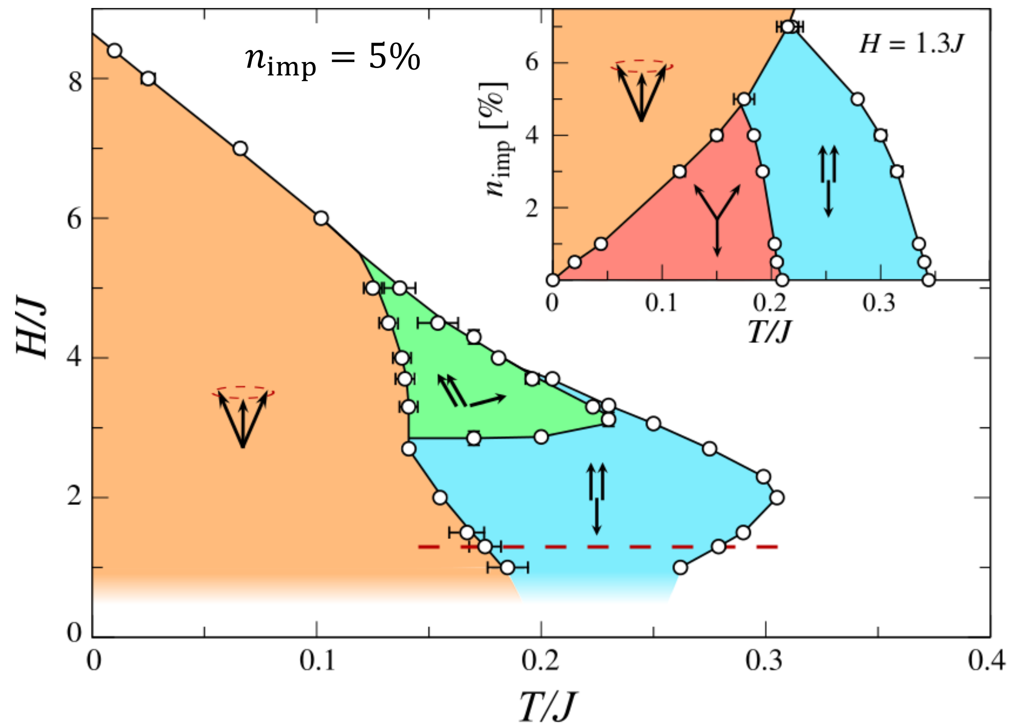
Conical phase
is a true GS at $T = 0$

Monte Carlo phase diagram

Periodic $L \times L$ romboedral cluster with $30 \leq L \leq 150$

All measured quantities are averaged over 100 random impurity configurations





V. Maryasin and M. Zhitomirsky, PRL (2013)

$$\Delta\hat{H} = -\frac{T}{18}\sum_{\langle i,j\rangle}\left(\boldsymbol{S_i}\cdot\boldsymbol{S_j}\right)^2 \qquad \text{vs} \qquad \Delta\hat{H} = \frac{\delta J^2}{6J}\sum_{\langle i,j\rangle}\left(\boldsymbol{S_i}\cdot\boldsymbol{S_j}\right)^2$$

Outline

Pure Heisenberg TAFM

HTAFM with impurities

Numerical methods and results

Beyond isotropic classical model

- Quantum ObD
- Easy-plane TAFM
- $Er_2Ti_2O_7$

Impurities vs Quantum ObD

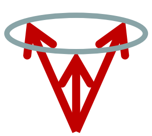
In all real systems quantum fluctuations must be taken into account

$$E = E_{\mathrm{GS}} + E_{\mathrm{th}} + E_{\mathrm{Q}} + E_{\mathrm{imp}}$$

$$E_Q = -\frac{JS}{24}\sum_{\langle i,j\rangle} \cos^2\theta_{ij} \qquad\qquad E_Q = \frac{\hbar}{2}\sum_k \omega_k$$

We compared selection energies at $H = 3J;\ \ T = 0;\ \ S = 5/2$

$$\Delta E = E_{\mathrm{cone}} - E_{\mathrm{uud}} = \Delta E_{\mathrm{Q}} + \Delta E_{\mathrm{imp}}$$

	$E_{\mathrm{Q}_{\mathrm{uud}}}$	$E_{\mathrm{imp}_{\mathrm{uud}}} = -H_{\mathrm{loc}} S n_{\mathrm{imp}}$
	$E_{\mathrm{Q}_{\mathrm{cone}}}$	$E_{\mathrm{imp}_{\mathrm{cone}}} = -H_{\mathrm{loc}} S n_{\mathrm{imp}} - JS^2 \alpha n_{\mathrm{imp}}$

$\alpha \simeq 0.65$ – from numerics

$\Delta E_{\mathrm{Q}} = 0.065 JS$ Chubukov, Golosov, JPCM (1991)

$n_{\mathrm{imp}} \sim 4\%$

Easy plane TAFM

$$\hat{H} = J\sum_{\langle i,j\rangle}\mathbf{S}^{\perp}{}_{i}\cdot\mathbf{S}^{\perp}{}_{j} - \mathbf{H}\sum\mathbf{S}^{\perp}{}_{i}$$

$$\Delta\hat{H} = -\frac{T}{18}\sum_{\langle i,j\rangle}\left(\boldsymbol{S_i}\cdot\boldsymbol{S_j}\right)^2 \quad \text{VS} \quad \Delta\hat{H} = \frac{\delta J^2}{3J}\sum_{\langle i,j\rangle}\left(\boldsymbol{S_i}\cdot\boldsymbol{S_j}\right)^2$$

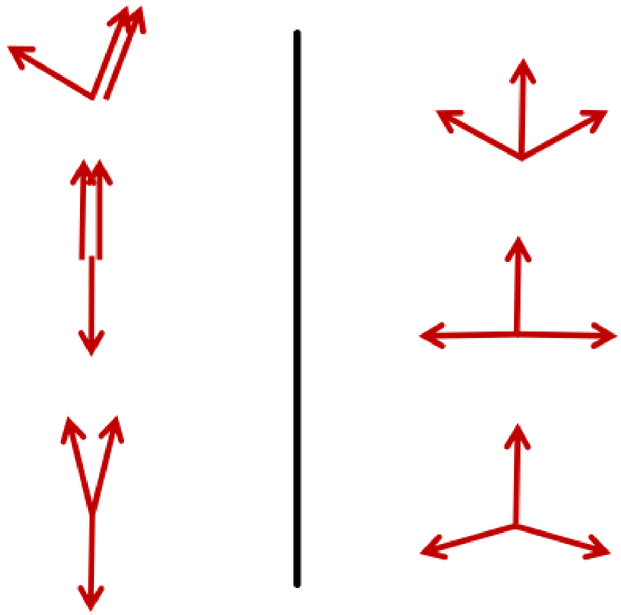
Thermal and quantum fluctuations stabilize the same phases

Pure phase diagram similar to Heisenberg model

Order by structural disorder stabilizes fan phase with no intermediate 1/3 uud plateau.

Hope for $\mathrm{Ba_3CoSb_2O_9}$

Thermal | Structural



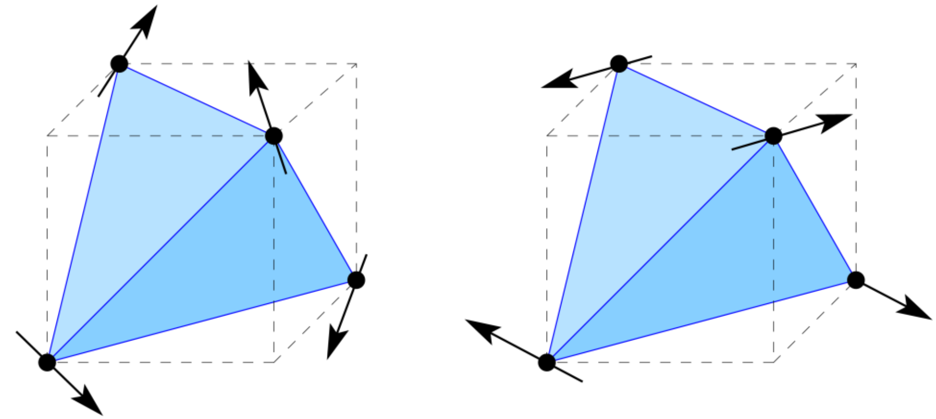
$Er_2Ti_2O_7$

Work in progress – impurities in frustrated XY - anisotropic pyrochlores.

$$\hat{H} = \sum_{\langle ij \rangle} J_\perp \mathbf{S}_i{}^\perp \cdot \mathbf{S}_i{}^\perp + J_\perp{}^a (\mathbf{S}_i{}^\perp \cdot \mathbf{r}_{ij})(\mathbf{S}_j{}^\perp \cdot \mathbf{r}_{ij}) + J_{zz} S_i{}^z S_j{}^z + J_{z\perp} \left[S_i{}^z (\mathbf{S}_j{}^\perp \cdot \mathbf{r}_{ij}) + S_j{}^z (\mathbf{S}_i{}^\perp \cdot \mathbf{r}_{ij}) \right]$$

For $J_\perp{}^a > 0$ an ordered state belongs to a 2-component $E(\Gamma_5)$ irrep of T_d

$$\mathbf{S}_i = \hat{\mathbf{x}}_i \cos\varphi + \hat{\mathbf{y}}_i \sin\varphi$$



$m_{3z^2-r^2}$ or $\hat{\mathbf{x}}$ $\quad$ $m_{x^2-y^2}$ or $\hat{\mathbf{y}}$

Experimentally detected that $Er_2Ti_2O_7$ orders at 1.2K into a $m_{3z^2-r^2}$ phase

Champion *et al*. PRB (2003)

Theories explain the $m_{3z^2-r^2}$ phase by thermal and quantum ObD

Zhitomirsky *et al.* PRL (2012)
Savary *et al.* PRL (2012)

Order by structural disorder in $Er_2Ti_2O_7$

MC simulations: classical pyrochlore with $J_\perp{}^a = 0.5\, J_\perp$ and $n_{\rm imp} = 5\%$ of vacancies

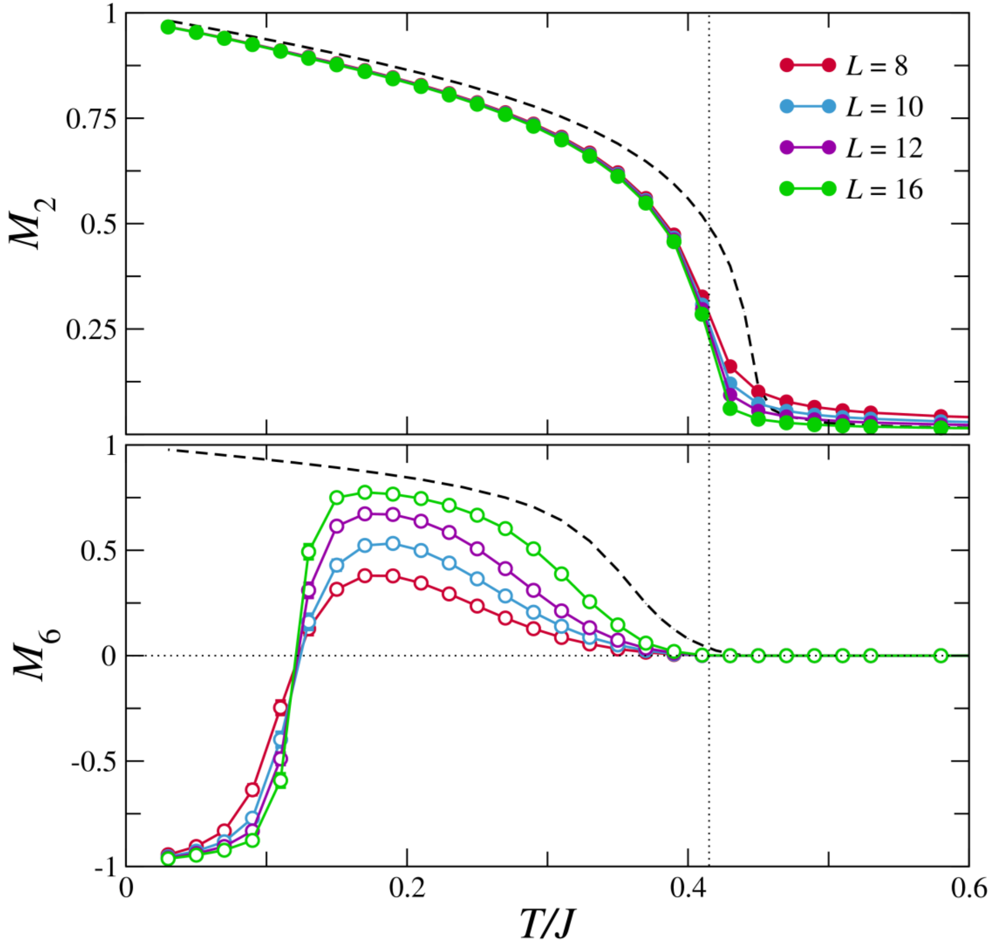
M_2 - AFM order parameter

$$M_2 = M_x{}^2 + M_y{}^2$$

M_6 - distinguishes $m_{\rm 3z^2-r^2}$ from $m_{\rm x^2-y^2}$ phase

$$M_6 \sim Re\left(M_x + iM_y\right)^6$$

Vacancies in $Er_2Ti_2O_7$ compete with thermal ObD and favor $m_{\rm x^2-y^2}$ states.



Summary

- Effective negative biquadratic exchange, responsible for thermal ObD and a phase diagram of the pure HTAFM

- Disorder-induced positive biquadratic exchange. It selects the least collinear states and competes with conventional ObD

- Phase diagram of HTAFM with vacancies

- Towards realistic models: order by structural disorder in easy-plane TAFM and $Er_2Ti_2O_7$

V. S. Maryasin and M. E. Zhitomirsky, PRL **111**, 247201 (2013)

Thank you for attention!

V. S. Maryasin and M. E. Zhitomirsky, PRL **111**, 247201 (2013)

INSTITUT NANOSCIENCES ET CRYOGÉNIE