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V. Spin dimer systems: ‚Kondo necklace‘ model, triplon BEC in dimer compounds ($TlCuCl_3$, Cs_2CuCl_4)

‚Mermin-Wagner theorem‘

An infinite d dimensional lattice of localized spins cannot have LRO at any finite temperature for $d < 3$ if the effective exchange interactions among spins are isotropic in spin space and of finite range.

- finite range: $$\bar{J} = \frac{1}{2\mathcal{N}} \sum_{ij} |J_{ij}| \, |\mathbf{x}_i - \mathbf{x}_j|^2 < \infty$$

- T=0 (ground state): even in 2D, 1D ground state may have LRO

- quasi - low D: small inter-chain (1D) or inter-plane (2D) interactions cause LRO at finite T

- phase transition without LRO: example: 2D Kosterlitz Thouless transition

I. The 2D classical xy-model and Kosterlitz Thouless transition

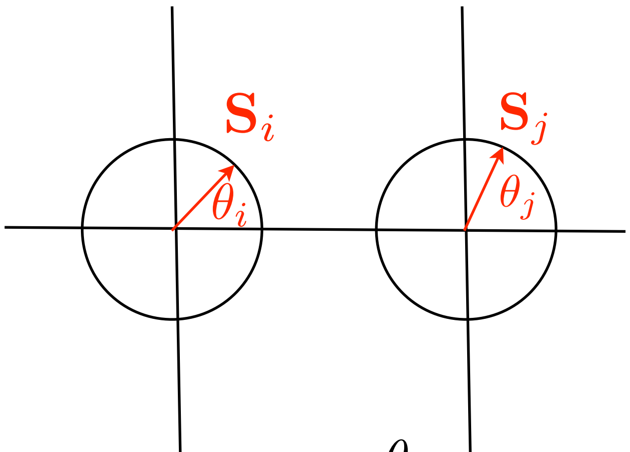
continuum limit of xy model

vortex configurations and interactions

thermodynamics of vortex-antivortex creation and KT transition

classical 2D xy or ‚rotor model'

$$H = -J \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j = -J \sum_{\langle i,j \rangle} \cos(\theta_i - \theta_j)$$



$$\mathbf{S}_i = \begin{pmatrix} \cos\theta_i \\ \sin\theta_i \end{pmatrix}$$

continuum approximation

$$H = E_0 + \frac{J}{2} \int d\mathbf{r} (\nabla \theta)^2.$$

$J \equiv \rho_s$ ‚spin stiffness'

$$S_+ = S_x + i S_y = e^{i\theta}$$

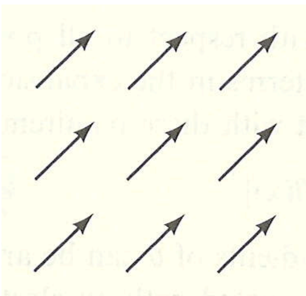
analogy to superfluidity

equilibrium configuration

$$\frac{\delta H}{\delta \theta(\mathbf{r})} = 0 \;\Rightarrow\; \nabla^2 \theta(\mathbf{r}) = 0$$

regular solution:

$$\theta(\mathbf{r}) = \theta_0$$



fluctuations (spin waves)

$$\theta(\mathbf{r}) = \theta_0 + \delta\theta(\mathbf{r})$$

$$\omega_{\mathbf{q}} = \rho_s \mathbf{k}^2$$

thermodynamics from spin fluctuations

symmetry breaking?

$$\langle \mathbf{S}_x \rangle = \frac{1}{Z} \int D\theta \cos\theta\left(\mathbf{r}\right) \exp\left[-\beta \frac{J}{2} \int d\mathbf{r} \left(\nabla\theta\left(\mathbf{r}\right)\right)^2\right] = \exp\left(-\frac{\mathcal{S}_d}{2Ja^{2-d}} AT\right)$$

d=2 $\quad A = ln(L/a) \quad \langle \mathbf{S} \rangle_T \to 0 \quad$ no LRO !

correlation function

$$\langle \mathbf{S}(\mathbf{r})\mathbf{S}(0) \rangle = \langle \cos(\theta(\mathbf{r})) \cos(\theta(0)) \rangle$$

$$\langle \mathbf{S}(\mathbf{r})\mathbf{S}(0) \rangle \simeq \begin{cases} e^{-\mathrm{const}.T} & \text{for } d > 2 \\ \boxed{\left(\frac{r}{L}\right)^{-\eta} \qquad \text{for } d = 2} \\ \exp(-\frac{T}{2Ja} r) & \text{for } d = 1 \end{cases} \qquad \eta = \frac{T}{2\pi J}$$

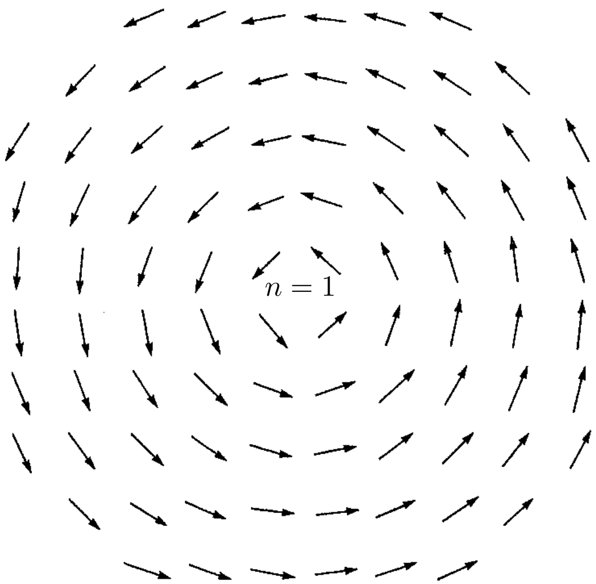
algebraic correlations at all temperatures: QLRO

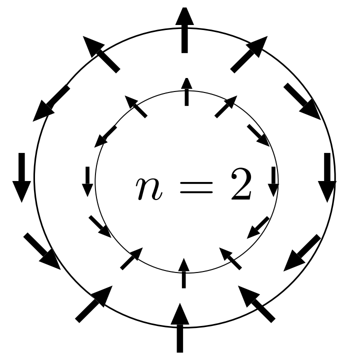
however: singular phase configurations have been neglected !

singular (vortex) solution:

$$\oint \nabla\theta(\mathbf{r})\cdot d\mathbf{l} = 2\pi n \qquad |\nabla\theta(r)| = n/r$$

n= ‚vortex charge'
topological nontrivial solution for n ≠ 0





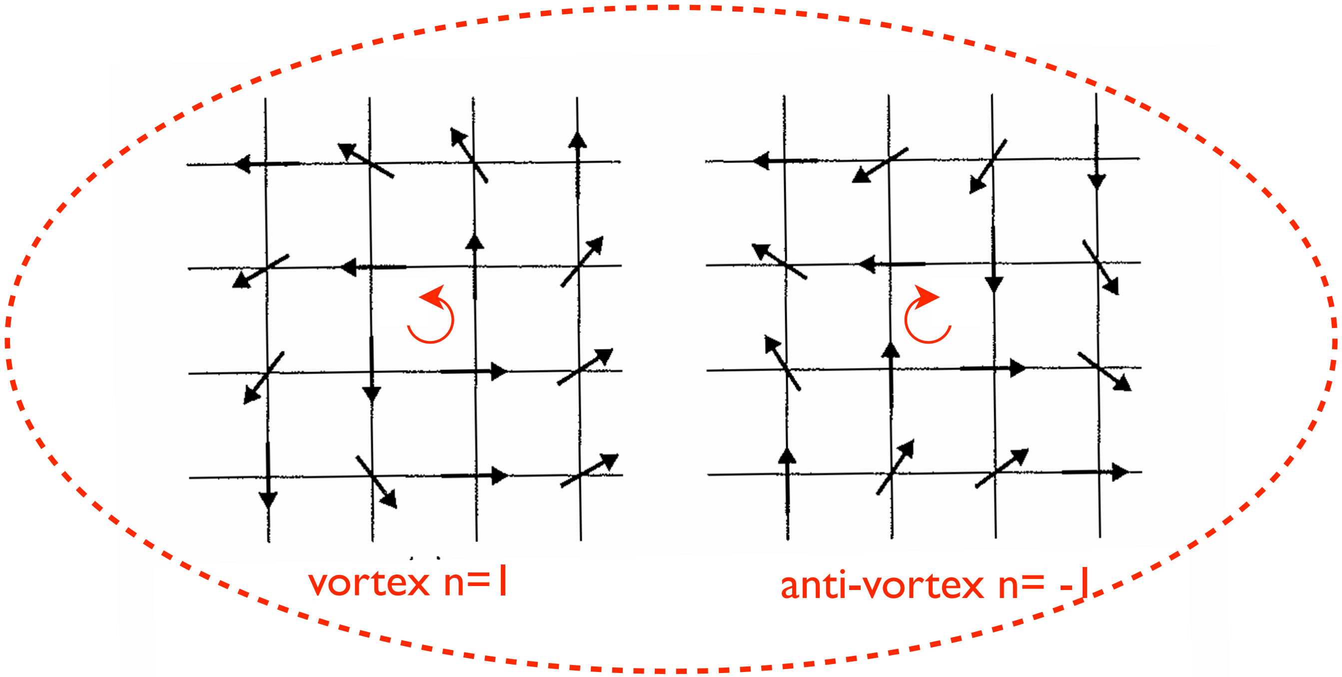
single vortex energy:

$$E_v = \frac{J}{2}\int d\mathbf{r}[\nabla\theta(\mathbf{r})]^2$$

$$= E_c(a) + \pi n^2 J \ln\left(\frac{L}{a}\right)$$

L=system size
a= core size

vortex-antivortex
pair:



$$\nabla\theta_v(\mathbf{r}) + \nabla\theta_a(\mathbf{r}) \to 0$$

interaction energy: $$E_{va} = 2E_c + E_1 \ln\left(\frac{|\mathbf{R}|}{a}\right)$$ ➽ v-a attraction

$$E_1 \sim \rho_s$$

Helmholtz free energy per vortex

$$F = E - TS$$

$$S = k_B \ln(L^2/a^2)$$

$$F = E_0 + (\pi J - 2k_B T)\ln(L/a) \begin{cases} < 0 & \text{for} \quad T > T_{KT} \\ > 0 & \text{for} \quad T < T_{KT} \end{cases} \qquad T_{KT} = \frac{\pi J}{2k_b}$$

screening by v-a pair creation

$$J \equiv \rho_s \;\rightarrow \rho_s^R$$

$$\rho_s^R = \rho_s - \frac{1}{2}\frac{\rho_s^2}{T}\lim_{k\to 0}\frac{\langle \hat{n}(\mathbf{k})\hat{n}(-\mathbf{k})\rangle)}{k^2}$$

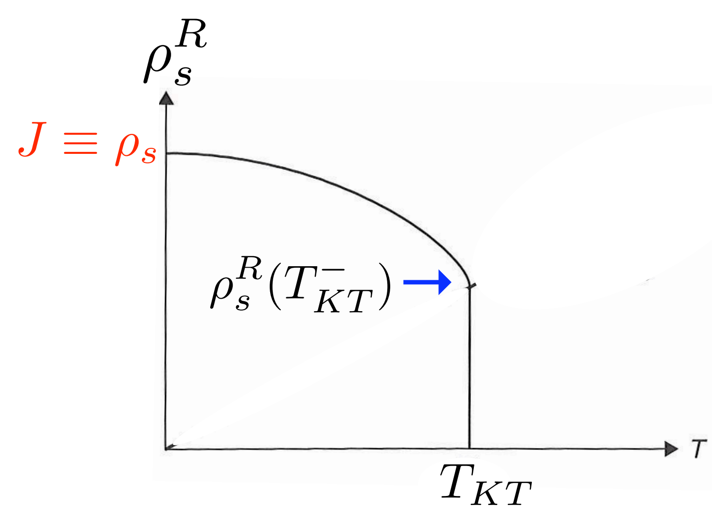
n(k) = vortex density

Kosterlitz-Thouless temperature: v-a unbinding

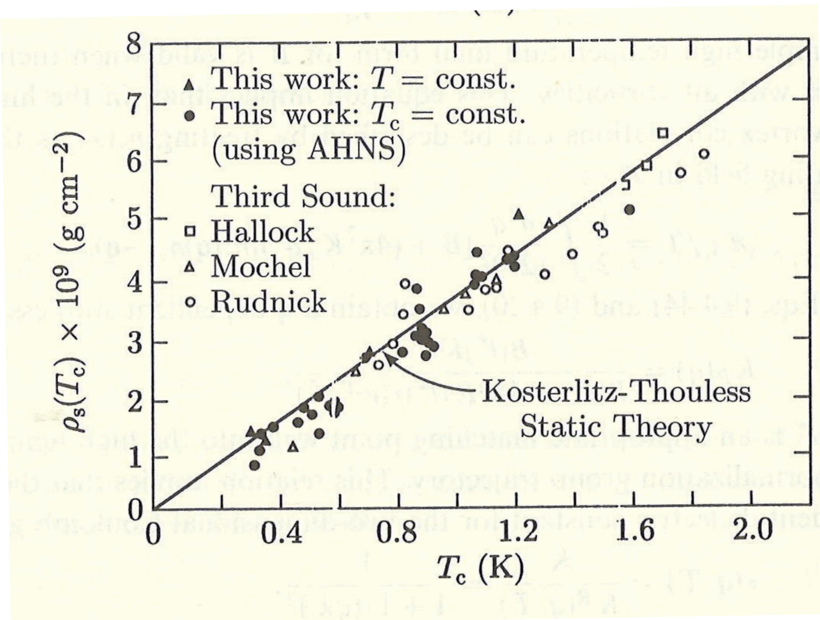
$$T_{KT} = \frac{\pi \rho_s^R(T_{KT}^-)}{2k_B}$$

universal KT ratio:

$$\frac{\rho_s^R(T_{KT}^-)}{k_B T_{KT}} = \frac{2}{\pi}$$



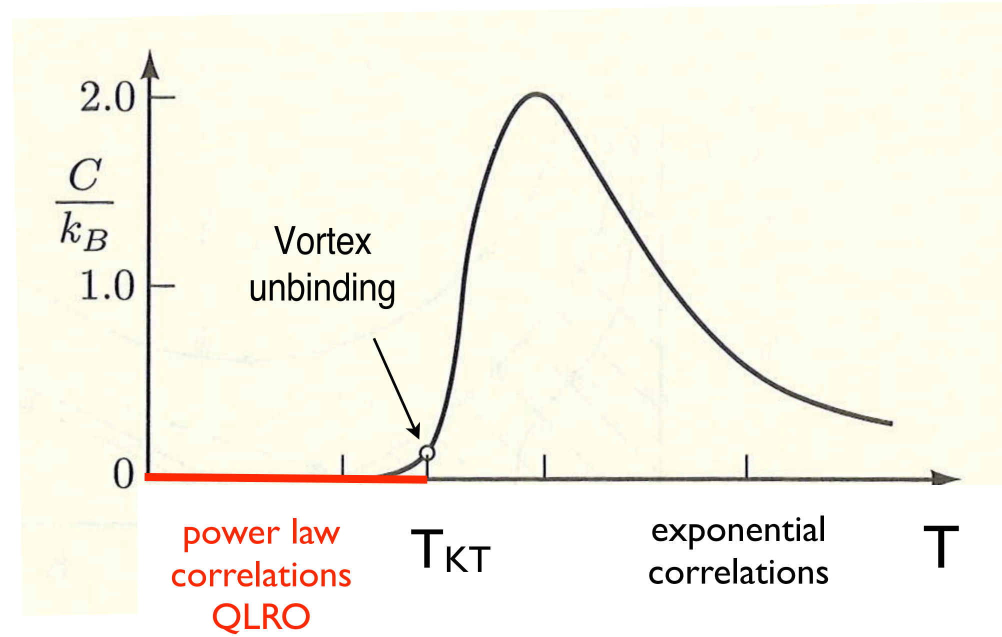
Universal KT ratio



$$\frac{\rho_s^R(T_c^-)}{k_B T_c} = \frac{2}{\pi}$$

exp. realisation:
superfluid ^{4}He thin films

Specific Heat



correlation length (T>T_{KT})

$$\xi(T) \sim \exp\left(\frac{const.}{(T - T_{KT})^{1/2}}\right)$$

$$\frac{F}{V} \sim \frac{T_{KT}}{\xi^2} \qquad C \sim \frac{\partial \xi^{-2}}{\partial T^2}$$

singularity cannot be seen

II. 2D square lattice J_1-J_2 quantum Heisenberg systems

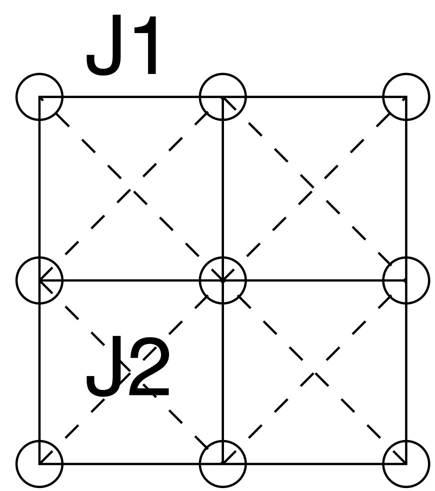
EPJB 38, 599 (04); PRB 76, 125113 (07);

PRB 77, 104441 (08)

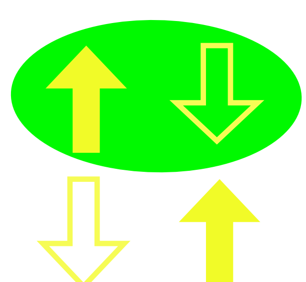
- Survey of J_1-J_2 vanadium compounds
- Zero-field phase diagram, methods to locate J_1-J_2 compounds (thermodynamic, neutron scatt.)
- Magnetisation and saturation field, role of quantum fluctuations
- Magnetocaloric effects: specific heat, adiabatic cooling rate

The frustrated 2D square lattice J_1-J_2 model

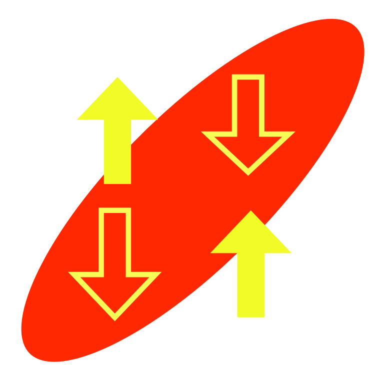
$$\mathcal{H} = J_1 \sum_{\langle ij \rangle_1} \mathbf{S}_i . \mathbf{S}_j + J_2 \sum_{\langle ik \rangle_2} \mathbf{S}_i . \mathbf{S}_k$$



J_1 alone favours
Néel order :



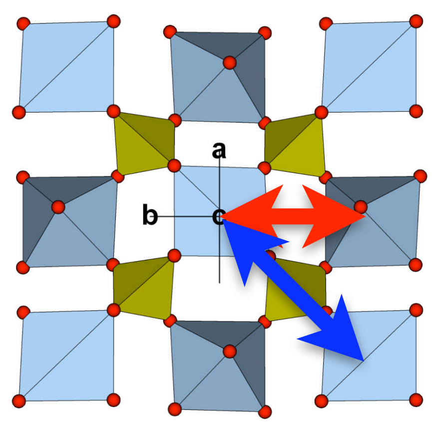
Introducing J_2 leads
to frustration :
„spin liquid state“?

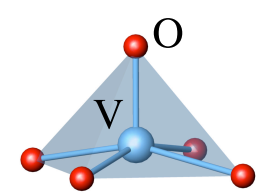


Old and new square lattice J_1-J_2 materials:

$Li_2VO(Si,Ge)O_4$	Melzi et al, PRL 85, 1318 (2000) Misguich et al, PRB 68, 113409 (2003)
$Pb_2VO(PO_4)_2$	Kaul et al, JMMM 272-276, 922 (2004)
$Zn_2VO(PO_4)_2$	Kini et al, J. Phys. Condens. Matter 18,1303 (2006)
$BaZnVO(PO_4)_2$ $SrZnVO(PO_4)_2$	Kaul, PhD thesis (2004)
$BaCdVO(PO_4)_2$	Nath, arXiv:0803.3535 (2008)

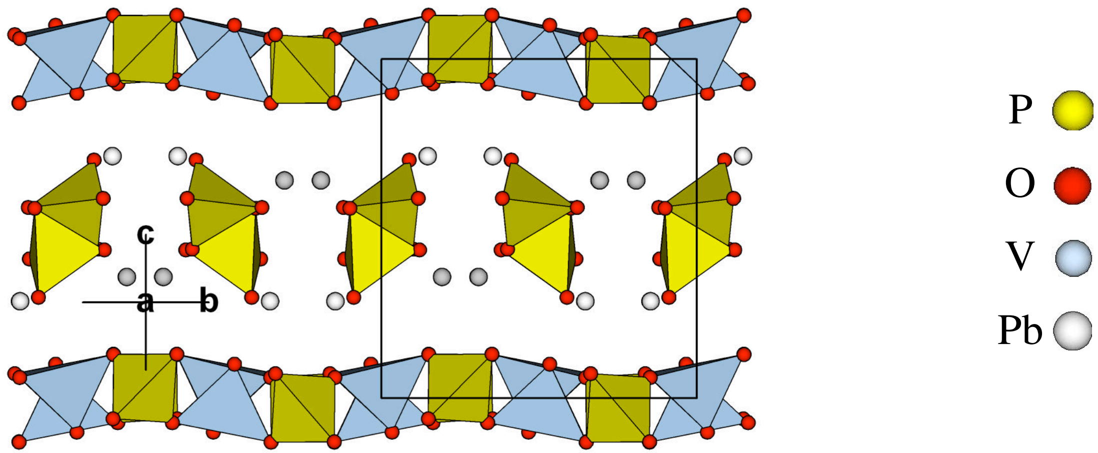
effective V-V exchange:
n.n. J_1
n.n.n. J_2



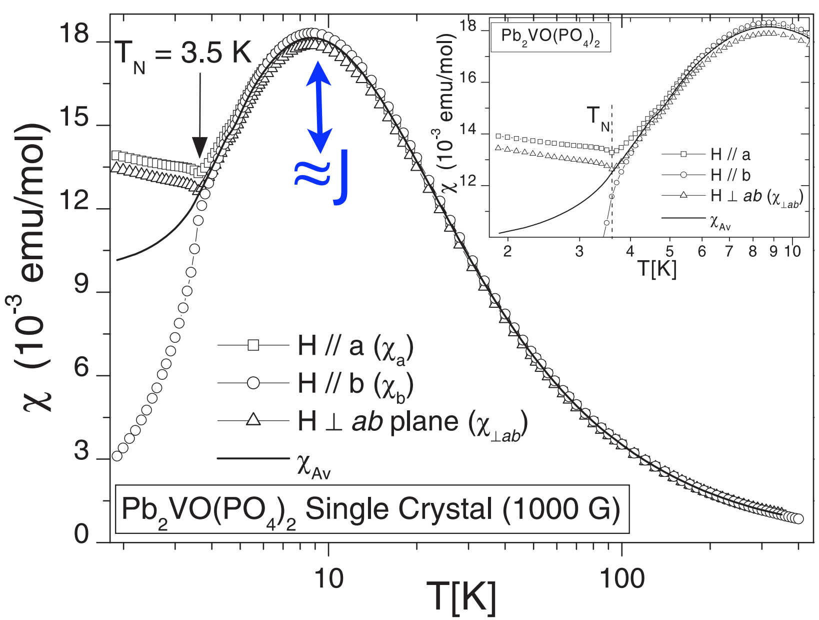


V^{4+}: $S = 1/2$

new 2D layered material: $Pb_2VO(PO_4)_2$



susceptibility



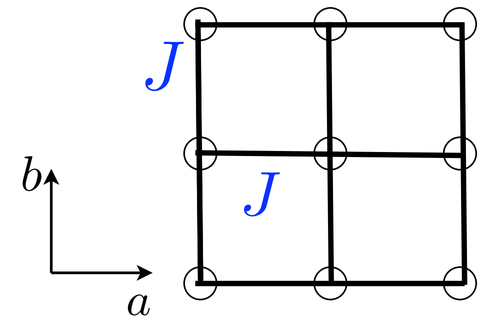
E. Kaul et al.,
J. Magn. Magn. Mat. **272–276** (2004) 922

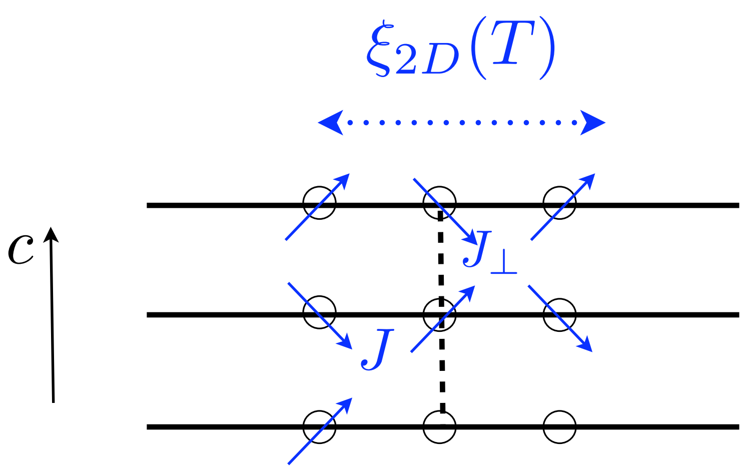
2D structure
but finite $T_N \sim J/3$??

quasi-2D and finite T_N for the simple HAF

$$J_2 = 0$$
$$J_1 \equiv J$$

$$H = J \sum_{ij\rangle} \mathbf{S}_i \cdot \mathbf{S}_j$$





2D correlation length diverges exponentially for T → 0

$$\frac{\xi_{2D}(T)}{a} \approx \left(\frac{J}{k_B T}\right) \exp\left(\frac{\pi J}{k_B T}\right)$$ QLRO

estimate for T_N:
thermal energy
≈ interlayer energy

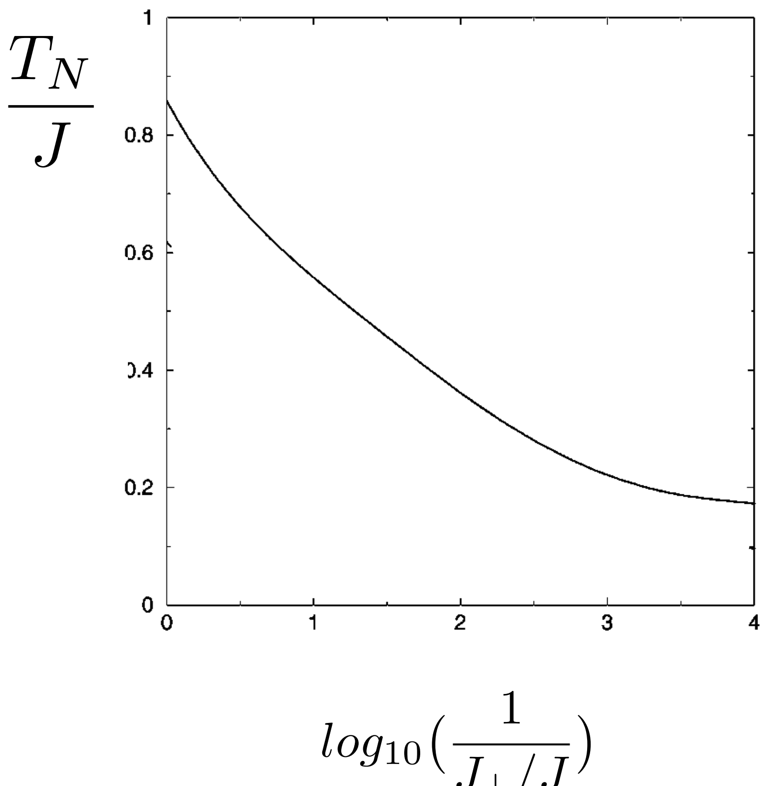
$$k_B T_N \approx \pi J_\perp S_e^2 \xi_{2D}^2 (T_N)$$

$$k_B T_N = \frac{\pi J}{c_0 + \ln\left(\frac{1}{J_\perp / J}\right)}$$

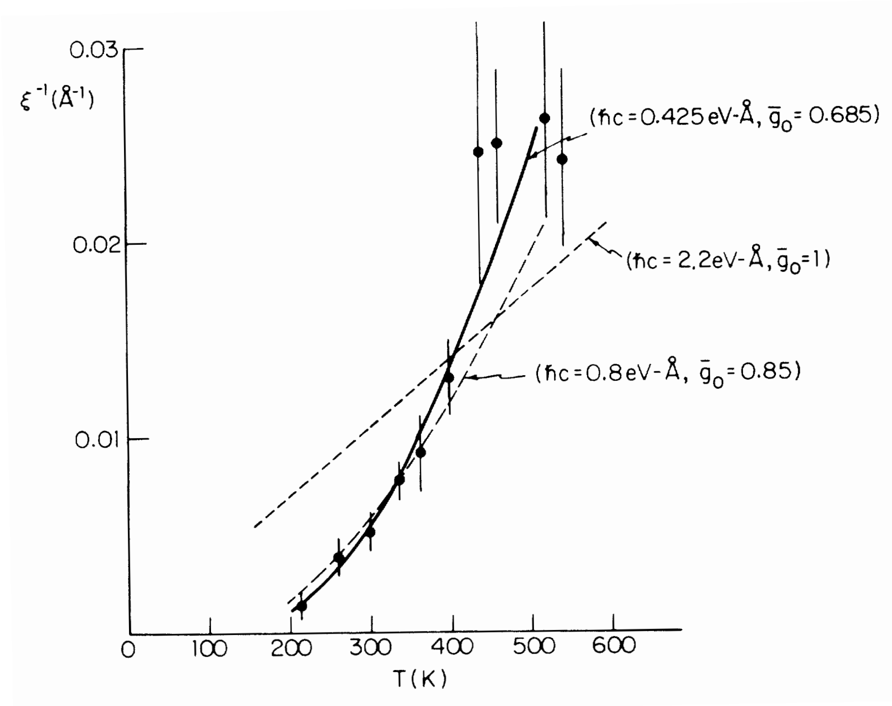
T_N of order J even for

$$J_\perp / J \ll 1$$

Siurakshina et al PRB 61,14601



best example: La_2CuO_4

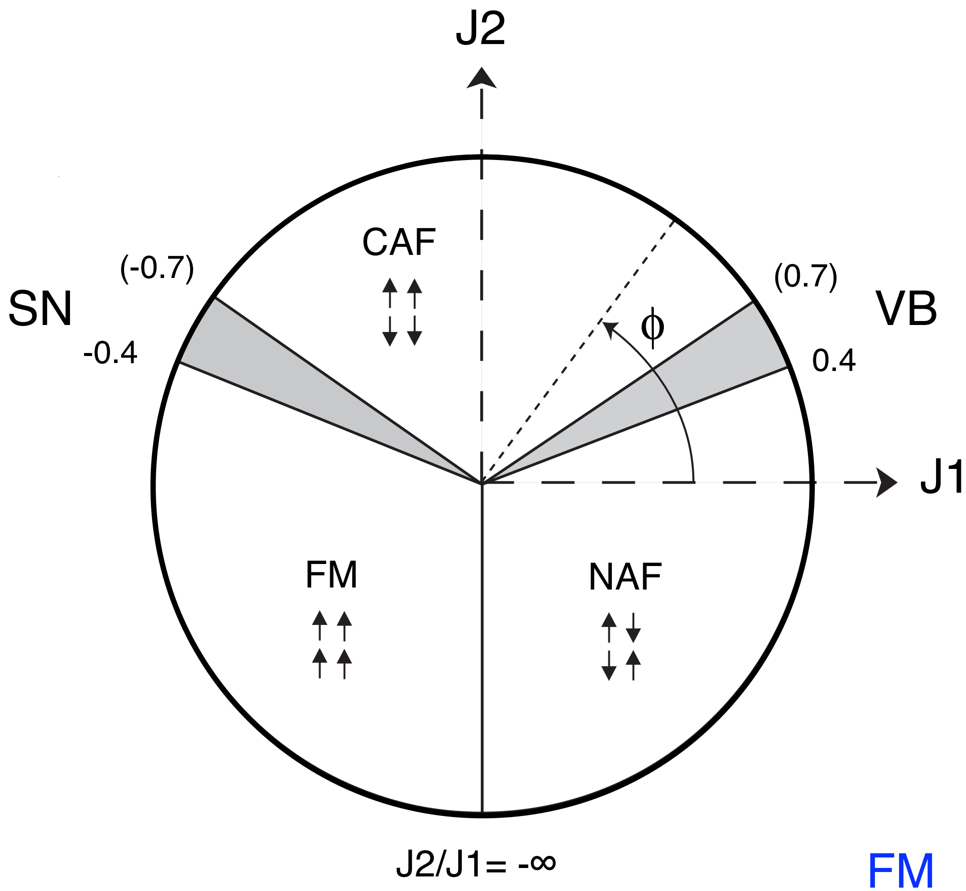


$$\frac{J_\perp}{J} < 10^{-4}$$ excellent quasi-2D

but 3D Neel order: $T_N \approx 200$ K

$$J \approx 1000 \text{ K}$$

signature of frustration: the J_1-J_2 phase diagram



$$\mathscr{H} = J_1 \sum_{\langle ij \rangle_1} \vec{S}_i \cdot \vec{S}_j + J_2 \sum_{\langle ik \rangle_2} \vec{S}_i \cdot \vec{S}_k$$

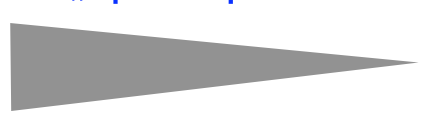
$$\boxed{J_c = \sqrt{J_1^2 + J_2^2}}$$

overall energy scale

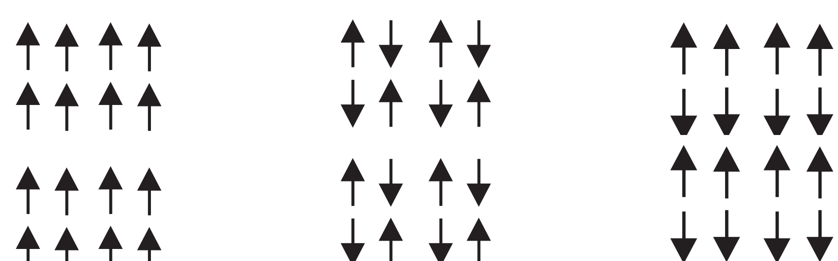
$$\boxed{\phi = \tan^{-1}(J_2/J_1)}$$

frustration parameter

„spin liquid“



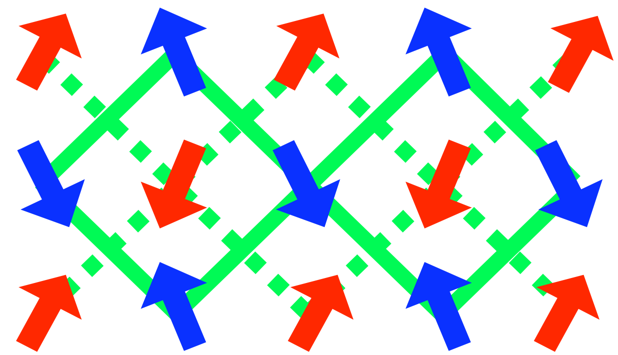
FM NAF CAF



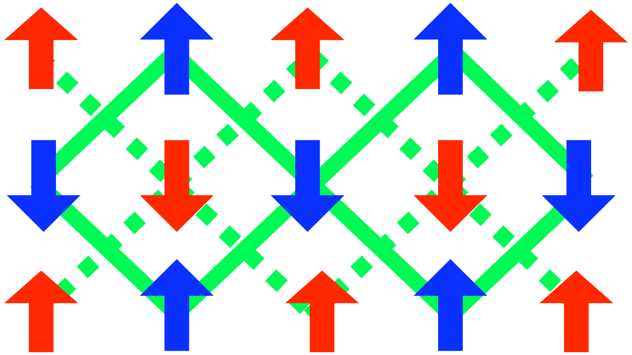
wave vector **Q**: $(0,0)$ (π,π) $(0,\pi)$

The CAF phase for $J_1 \approx 0$

$J_1 = 0$: $J_2 > 0$ favors
two independent
Néel sublattices :



Infinitesimal $|J_1|$:
quantum fluctuations
select **CAF** state



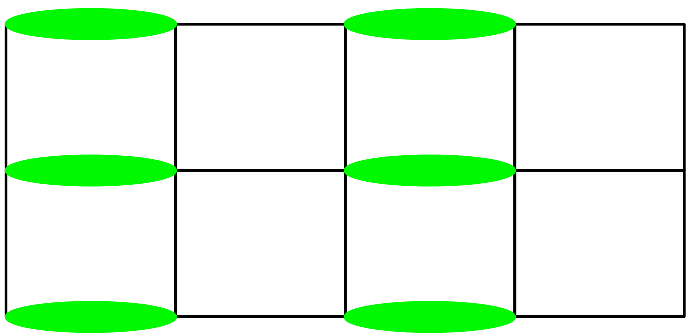
(Quantum) Order from (classical) disorder

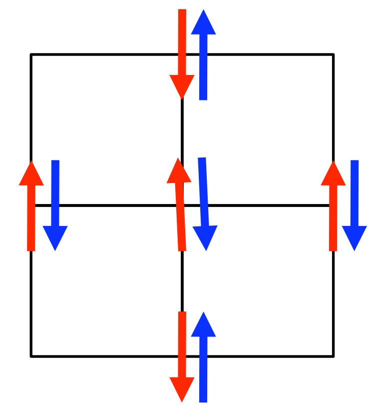
proposals for the **magnetically** disordered sectors

no true spin liquid, but hidden order

VB: $\boldsymbol{J_1 \approx 2J_2}$

SN: $\boldsymbol{J_1 \approx -2J_2}$





$\frac{1}{\sqrt{2}} \left| \uparrow\downarrow - \downarrow\uparrow \right\rangle =$ 

$$O^{\alpha\beta}(i,j) = \frac{1}{2}(S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha) - \frac{1}{3}\delta^{\alpha\beta}\langle \mathbf{S}_i \mathbf{S}_j \rangle$$

Valence Bond crystal of singlets

breaks translational symmetry

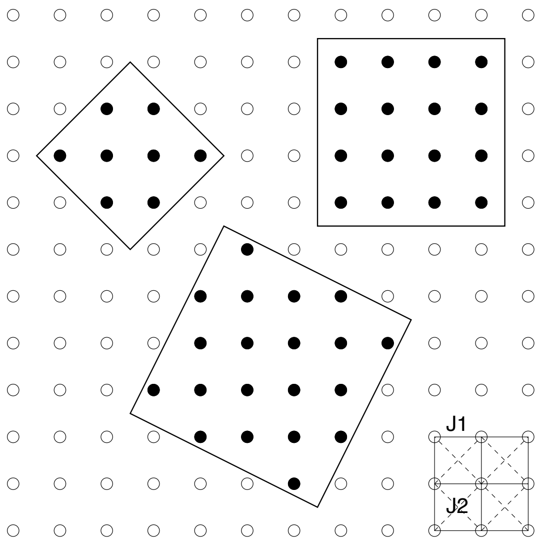
Spin Nematic state (nonlocal spin quadrupole)

breaks spin rotational symmetry

both preserve time reversal

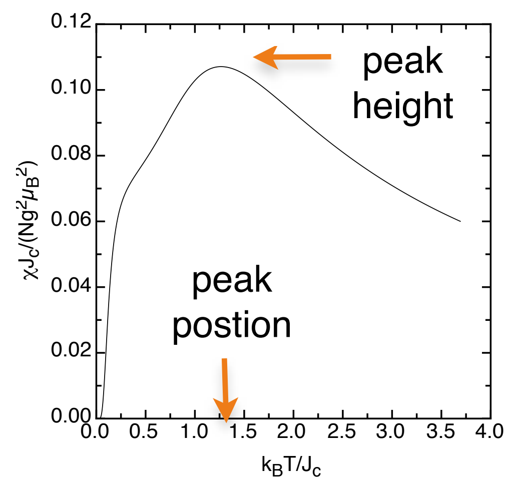
Thermodynamic quantities

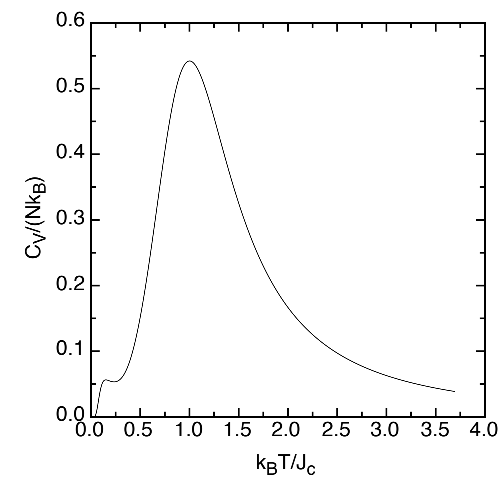
finite-T Lanczos method (FTLM):



$$\chi(T) = \frac{1}{T}\left\langle \left(S_z^{\text{tot}} - \langle S_z^{\text{tot}}\rangle\right)^2 \right\rangle$$

$$C_V(T) = \frac{1}{T^2}\left\langle \left(H - \langle H\rangle\right)^2 \right\rangle$$





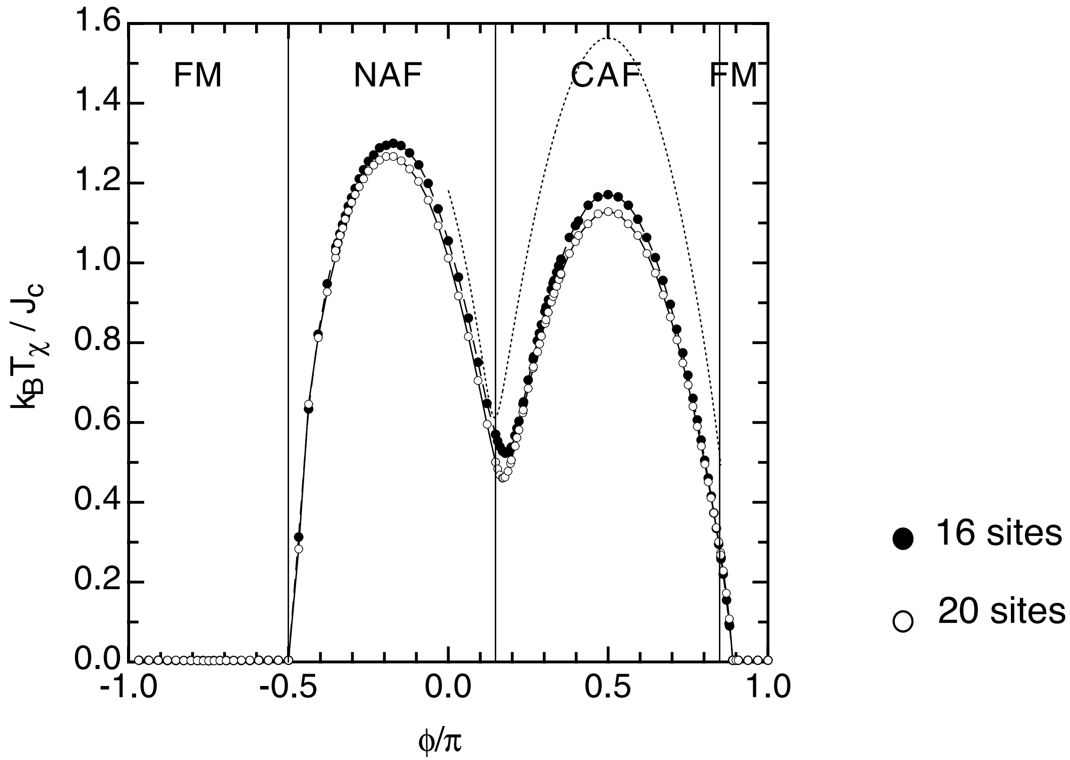
partition function for the cluster
generated from 500…1000
successive iterative diagonalizations:
correlation functions

Cluster size N = 16, 20, 24

$J_1 = 1.0$
$J_2 = -0.6$

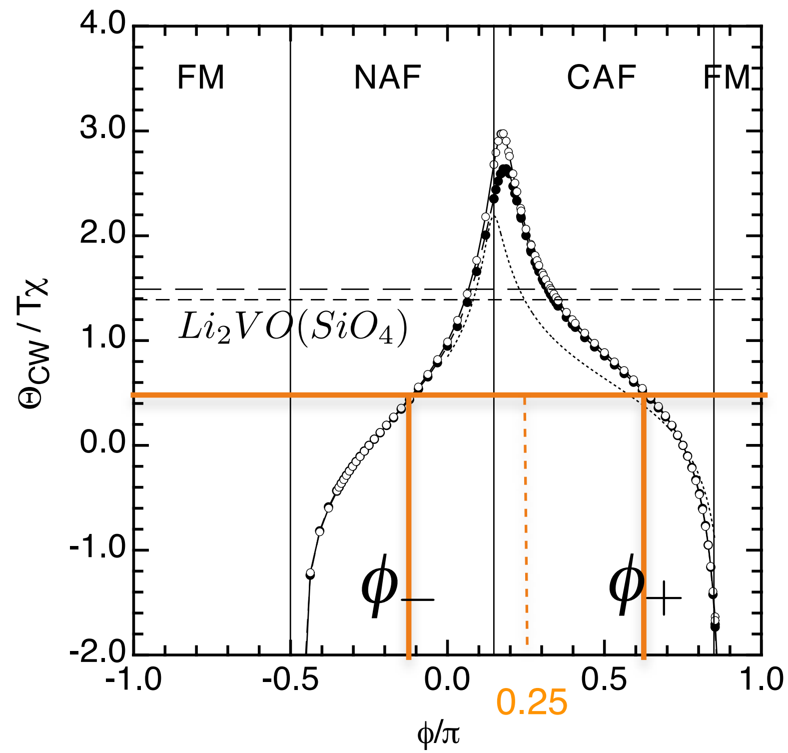
magnetic susceptibility

peak position T_χ



or: Θ_{CW}/T_χ

$\Theta_{CW} = J_1+J_2= J_c(\cos\phi+\sin\phi)$



$Pb_2VO(PO_4)_2$

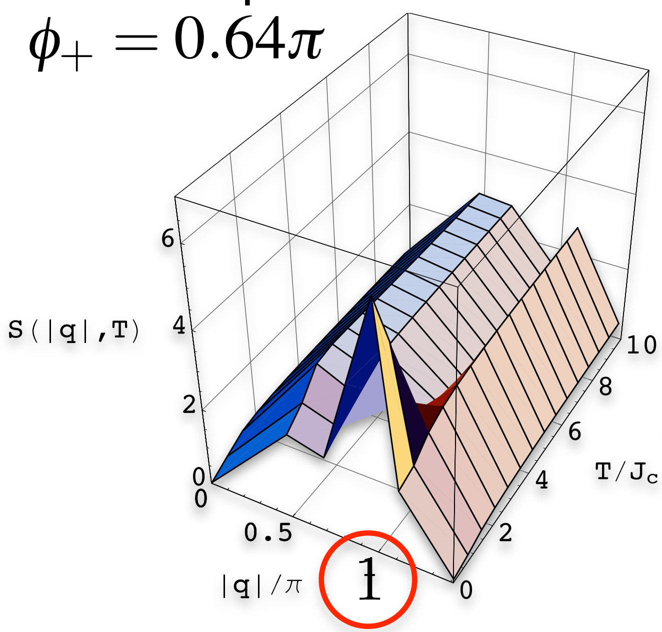
$\phi_\pm$ ambiguity for exchange ratio

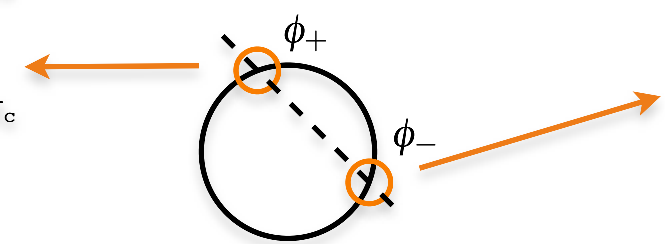
additional diagnostic tool: static spin structure factor

$$S(\mathbf{q},T) = \frac{1}{N}\sum_{i,j=1}^{N} e^{i\mathbf{q}(\mathbf{R}_i-\mathbf{R}_j)}\left\langle \mathbf{S}_i\mathbf{S}_j\right\rangle$$

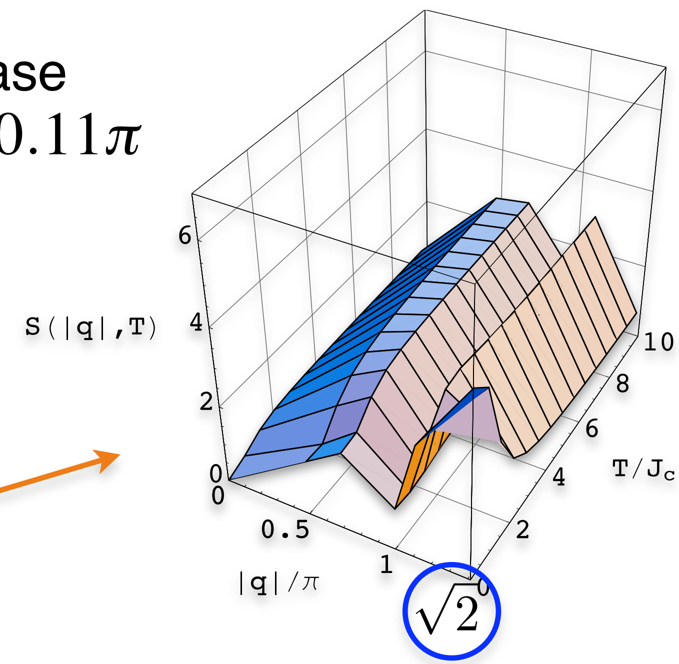
poly-averaged:

collinear phase
$\phi_+ = 0.64\pi$





Néel phase
$\phi_- = -0.11\pi$

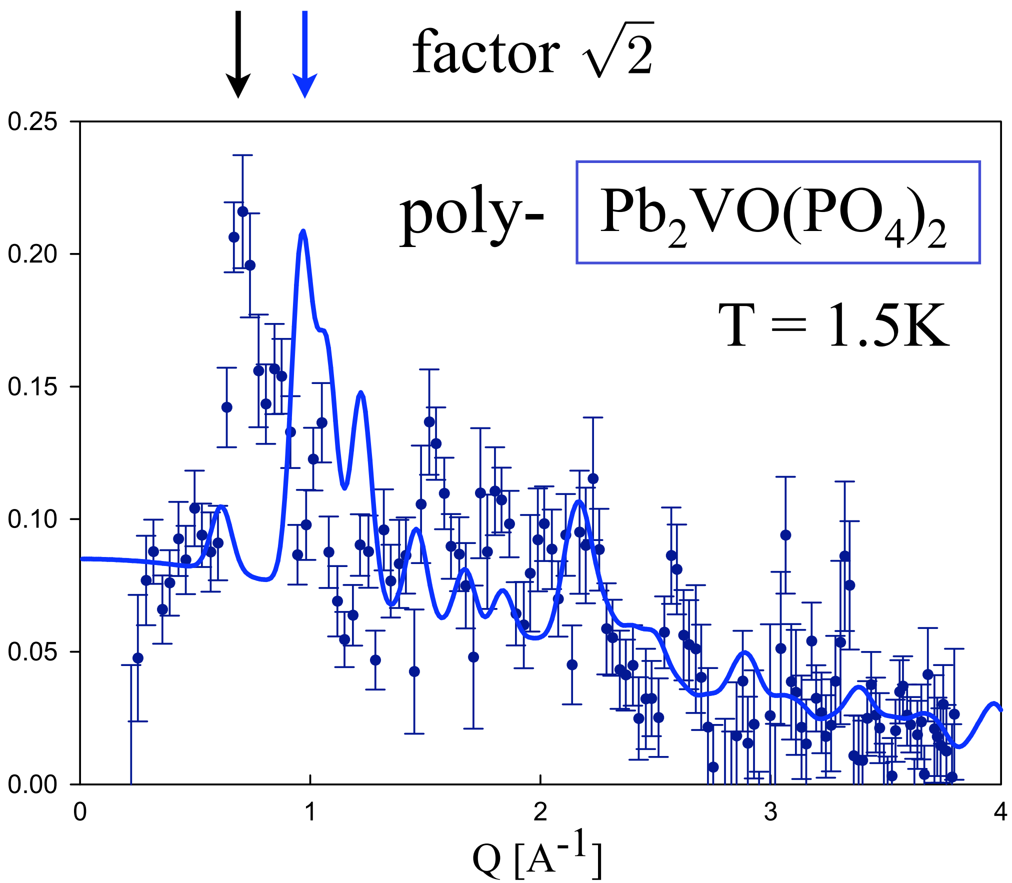


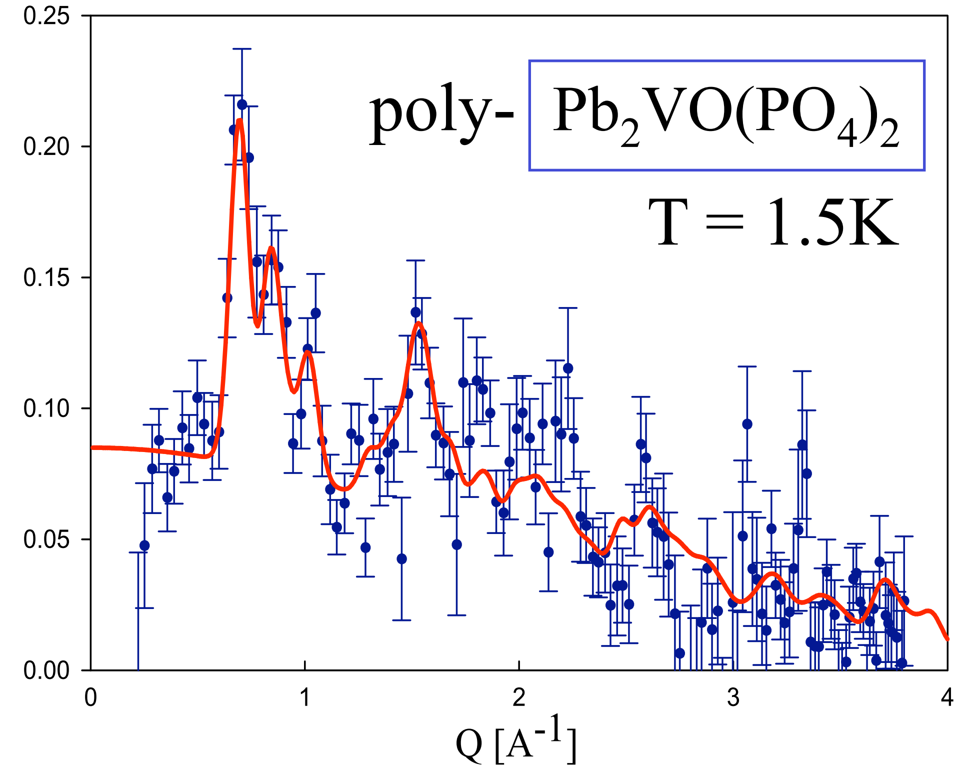
Ref.: N. Shannon et al., Eur. Phys. J. B **38** (2004) 599

diffuse neutron scattering can measure S(|**q**|, *T*) for $T > T_N$

proof of CAF ground state from S(Q) in polarized neutron diffraction

Skoulatos et al, JMMM 310, 2257 (2007)





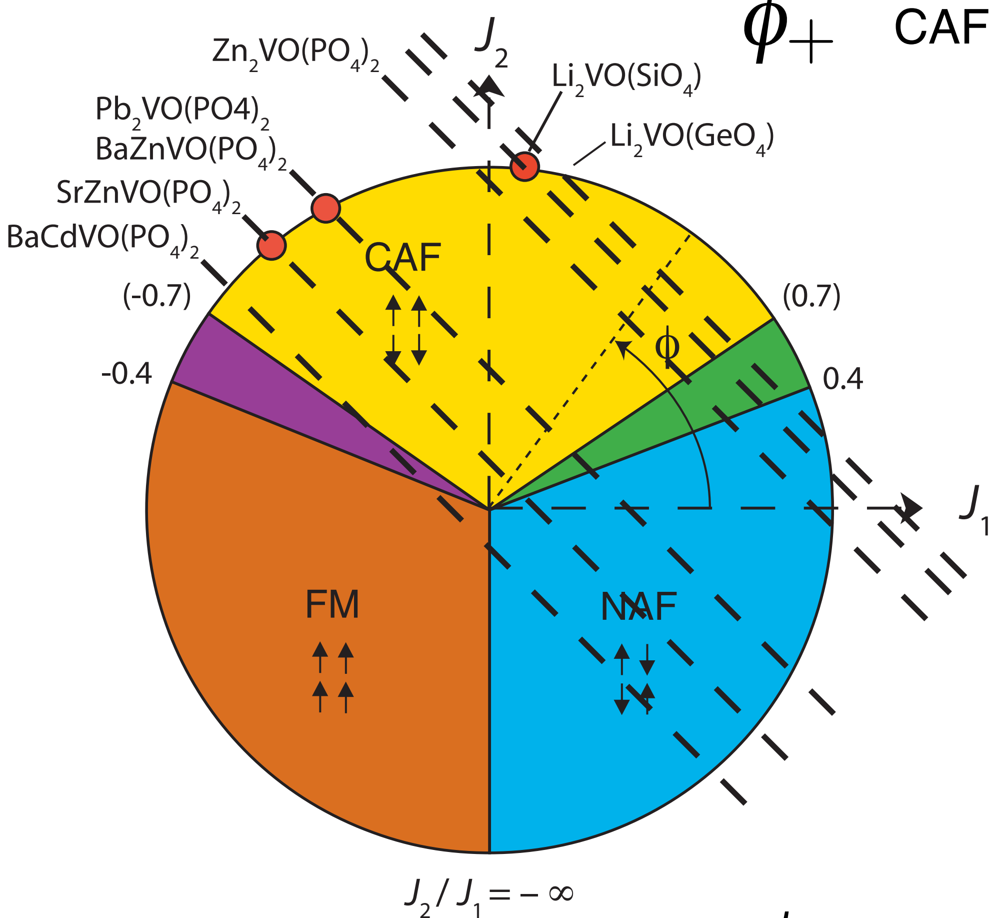
Fit with Néel model

$Q = (\pi,\pi)$

Fit with CAF model

$Q = (0,\pi)$

Compound location in J_1-J_2 plane



ϕ_+ CAF: ordering vector (π, 0)

ϕ_- NAF: ordering vector (π, π)

$J_c = \sqrt{J_1^2 + J_2^2}$ from

$$C_V(T \to \infty) = \frac{3}{8}\frac{J_c^2}{T^2} + \cdots$$

$\Theta_{CW} = J_1 + J_2$ from

$$\chi(T \to \infty) = \frac{S(S+1)}{3}\frac{1}{T}\left(1 + \frac{\Theta_{CW}}{T} + \cdots\right)$$

From $\chi(T)$, $C_V(T)$: Φ -determination is ambiguous

are there further methods of
diagnostics to discriminate $\Phi_{\pm}$?

investigate high-field properties:
magnetization + saturation fields