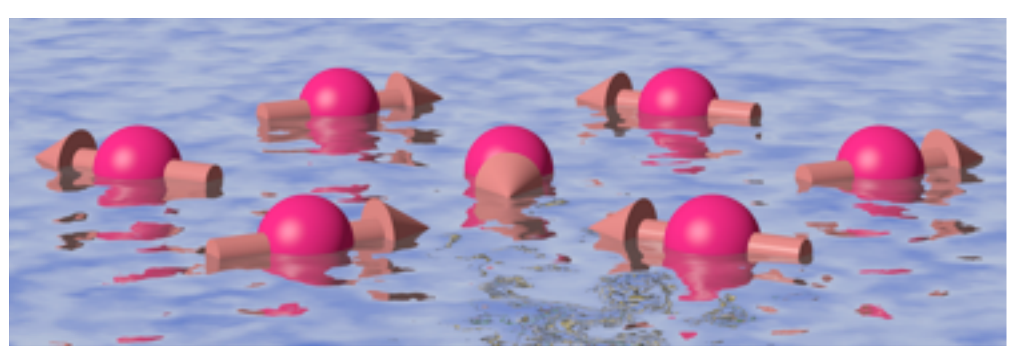
Triangular lattice antiferromagnet in magnetic field: ground states and excitations

Oleg Starykh, University of Utah

Jason Alicea, Caltech
Leon Balents, KITP
Andrey Chubukov, U Wisconsin



Outline

- motivation: Cs_2CuBr_4 , Cs_2CuCs_4
- Spin waves in non-collinear spin structures
- classical antiferromagnet in a field: entropic selection
  - spatial anisotropy - high-T stabilization of the plateau
- Quantum spins: zero-point fluctuations
  - Large-S analysis of interacting spin waves
- Approach from one dimension
  - sequence of plateaux and selection rules

★ (attempt at) Unification

VOLUME 86, NUMBER 7 PHYSICAL REVIEW LETTERS 12 FEBRUARY 2001

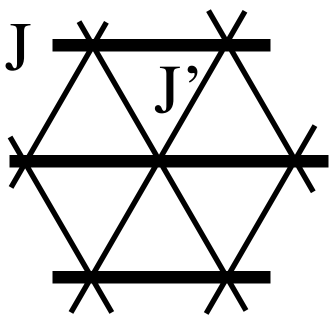
Experimental Realization of a 2D Fractional Quantum Spin Liquid

R. Coldea,[1,2] D. A. Tennant,[2,3] A. M. Tsvelik,[4] and Z. Tylczynski[5]

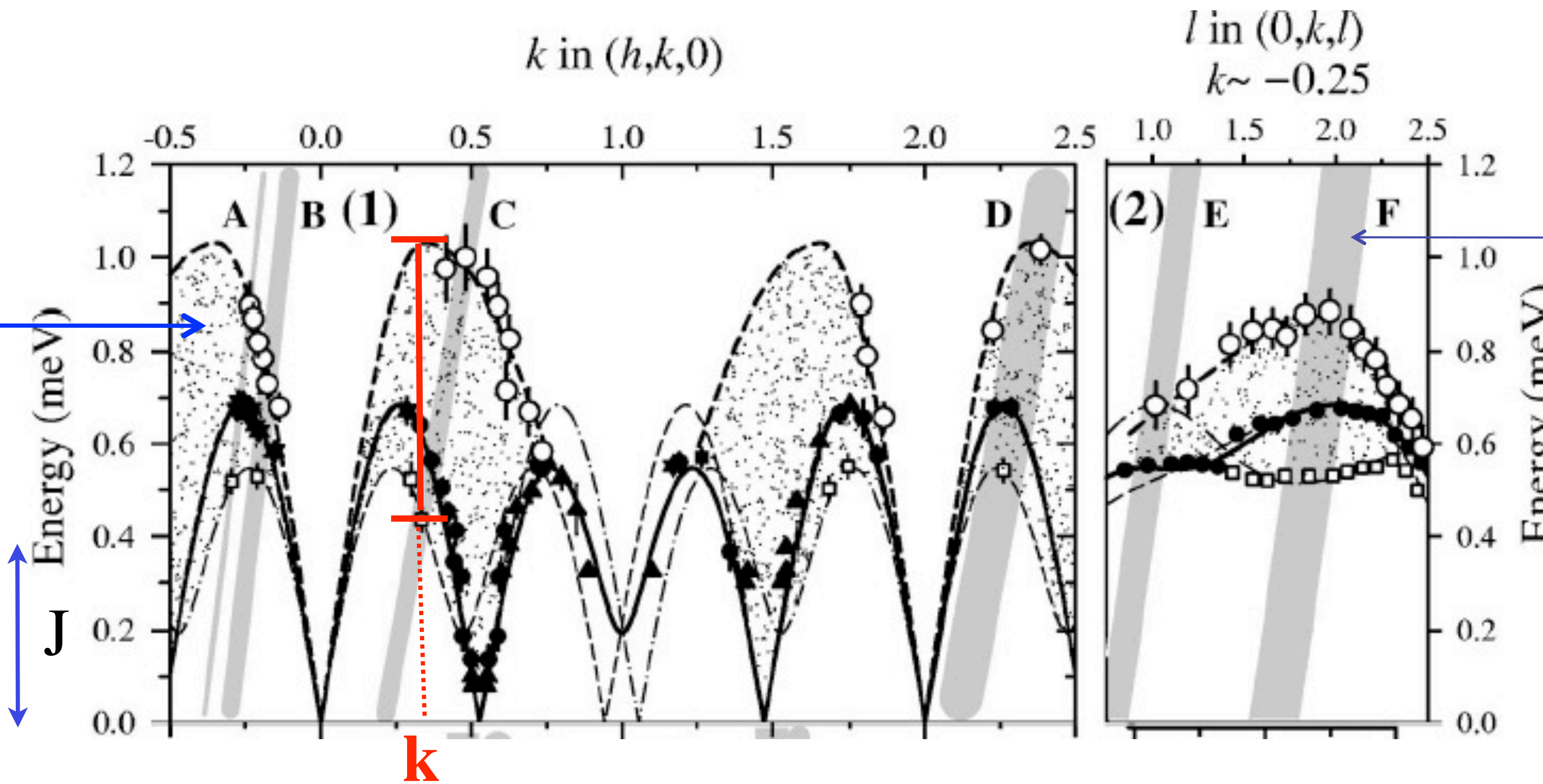
PHYSICAL REVIEW B 68, 134424 (2003)

Extended scattering continua characteristic of spin fractionalization in the two-dimensional frustrated quantum magnet Cs_2CuCl_4 observed by neutron scattering

R. Coldea,[1,2,3] D. A. Tennant,[1,3] and Z. Tylczynski[4]



Along the chain



transverse to chain

Cs_2CuCl_4
J'/J=0.34

Very unusual response: broad and strong continuum; spectral intensity varies strongly with 2d momentum (k_x, k_y)

VOLUME 86, NUMBER 7 PHYSICAL REVIEW LETTERS 12 FEBRUARY 2001

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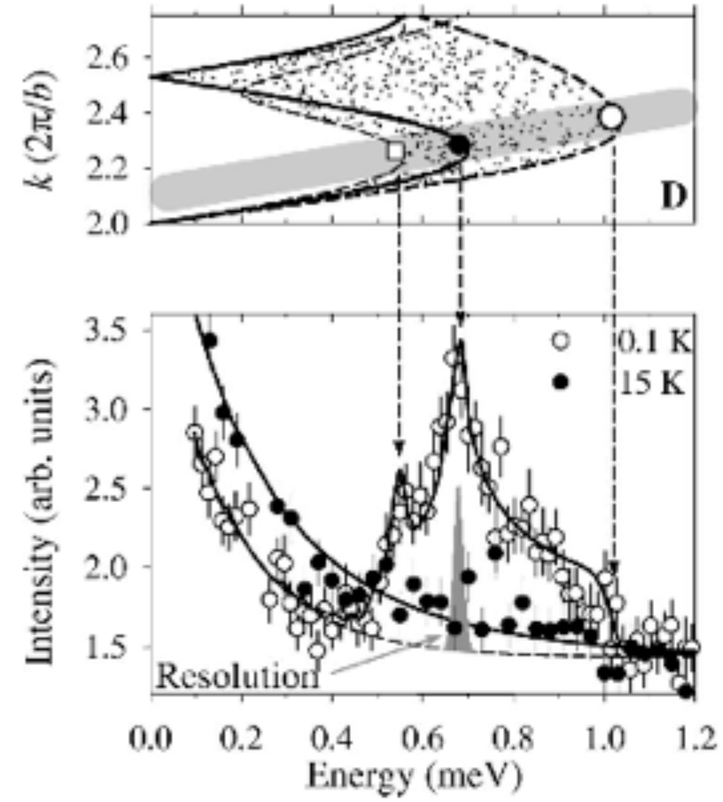

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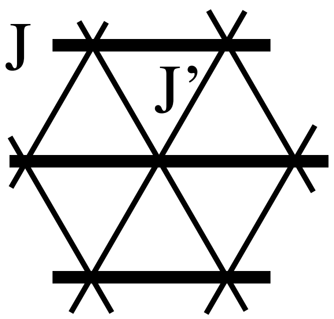

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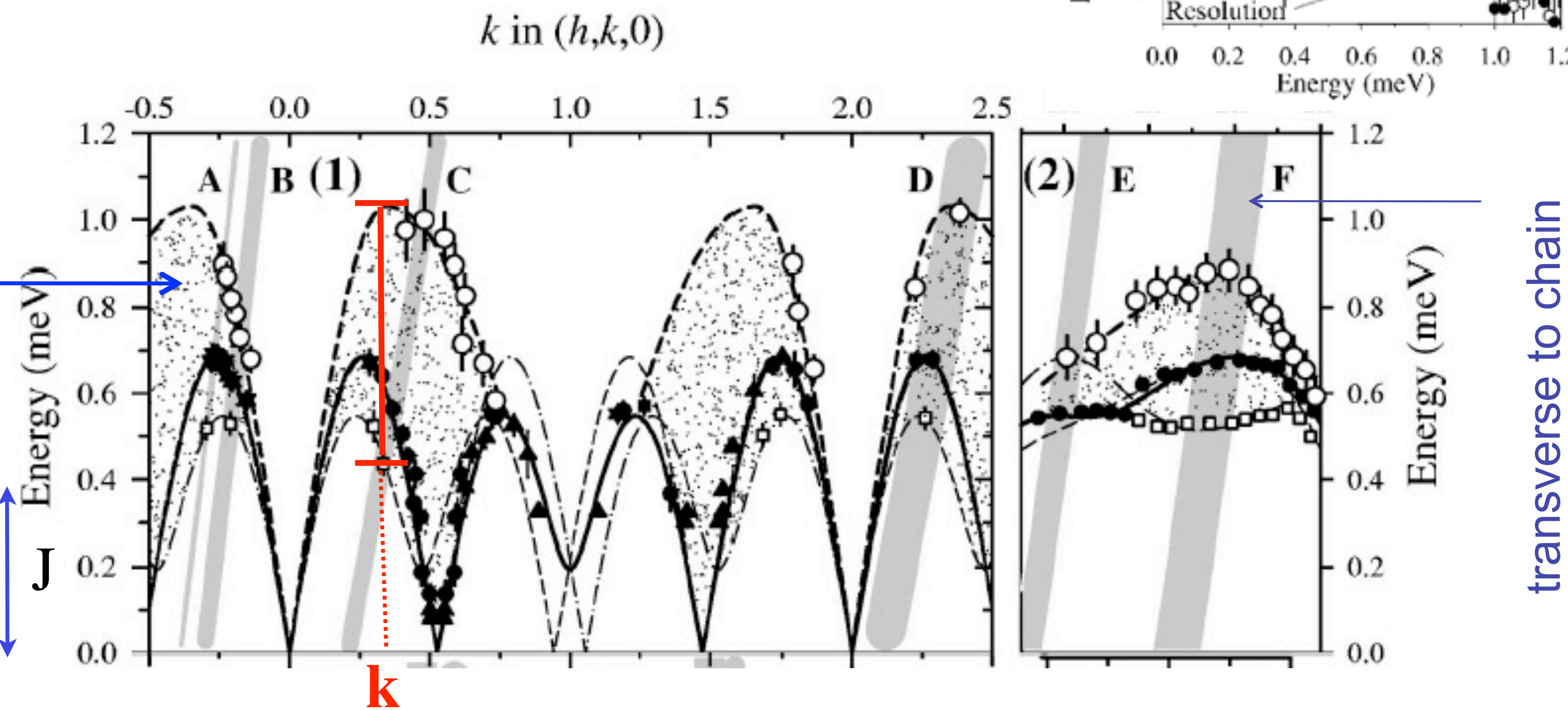


Cs_2CuCl_4

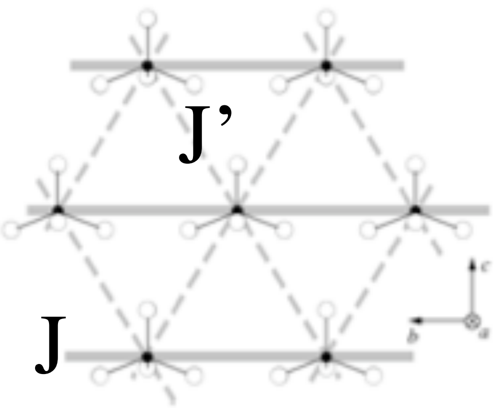
$J'/J=0.34$

Very unusual response: broad and strong continuum; spectral intensity varies strongly with 2d momentum (k_x, k_y)

M=1/3 magnetization plateau in Cs_2CuBr_4

★ Observed in Cs_2CuBr_4 (Ono 2004, Tsuji 2007) **J'/J = 0.75**
but not Cs_2CuCl_4 [**J'/J = 0.34**]

S=1/2



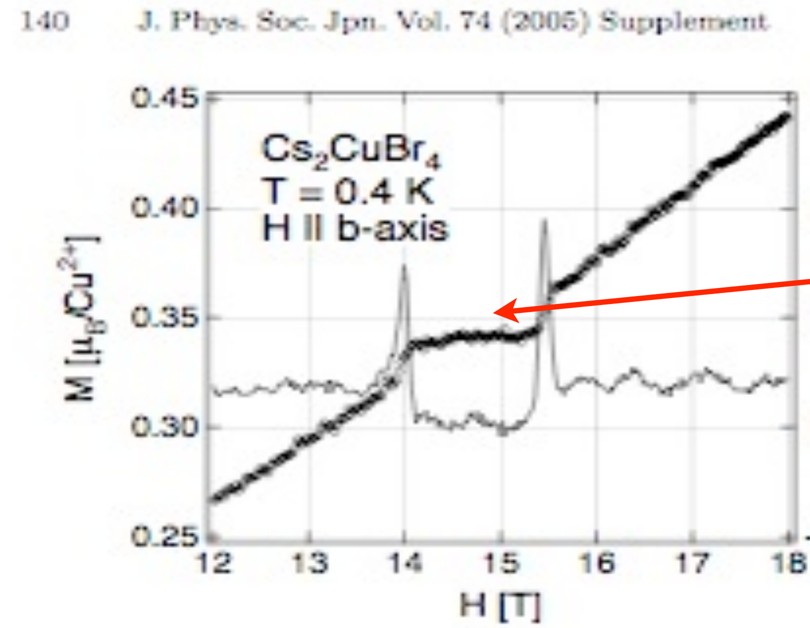
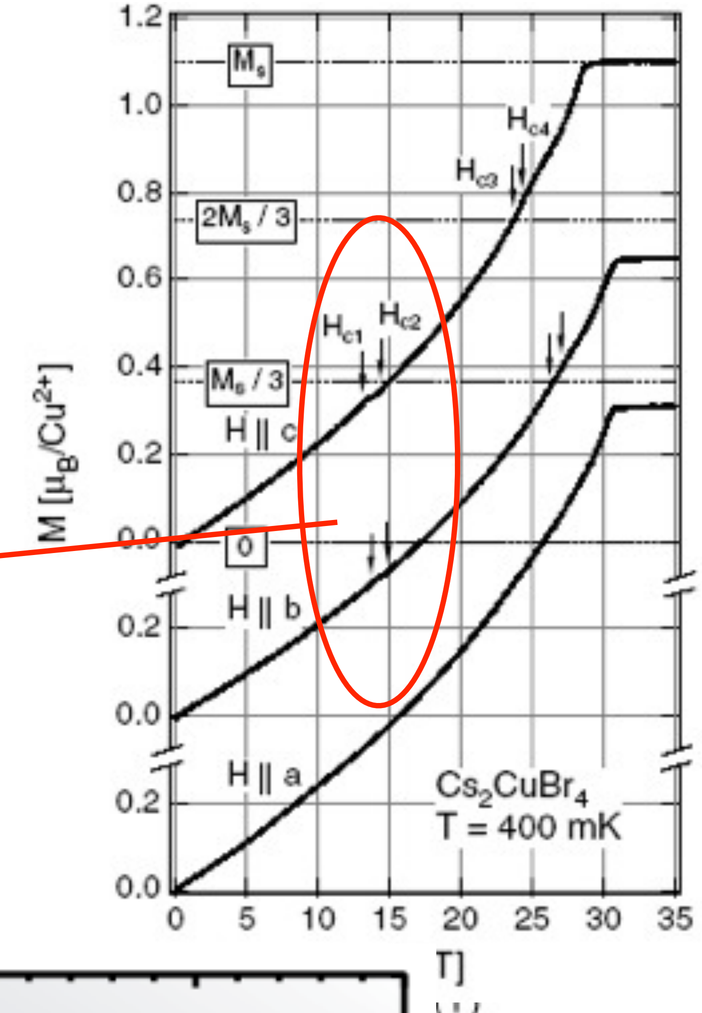
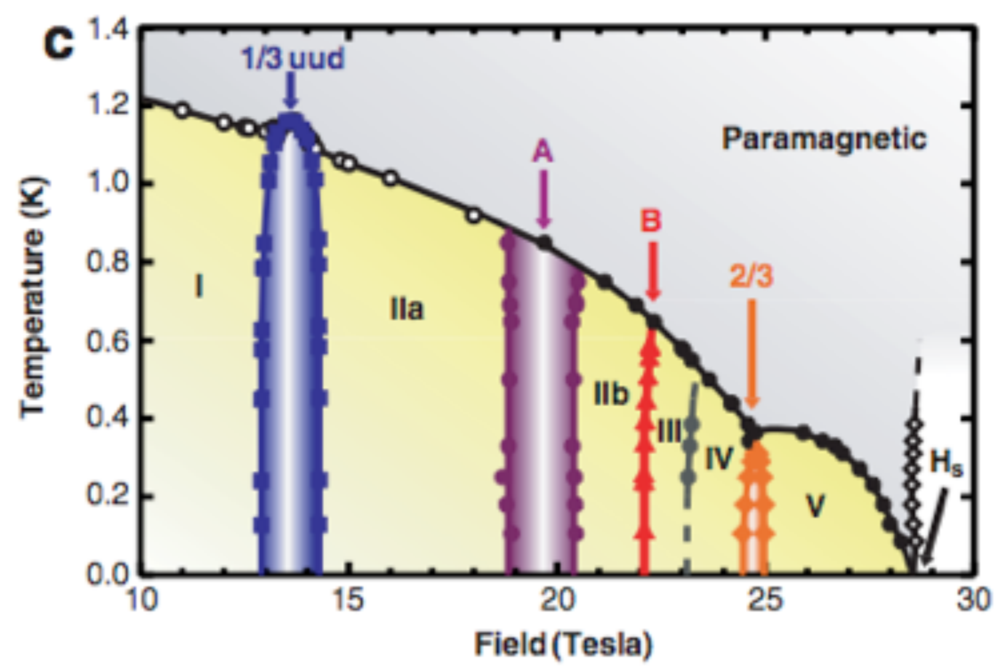


Fig. 8. The magn
measured at T =





★ first observation of "**up-up-down**" state
in spin-1/2 triangular lattice antiferromagnet

★ and **8** more **phases** (instead of 2 expected)!

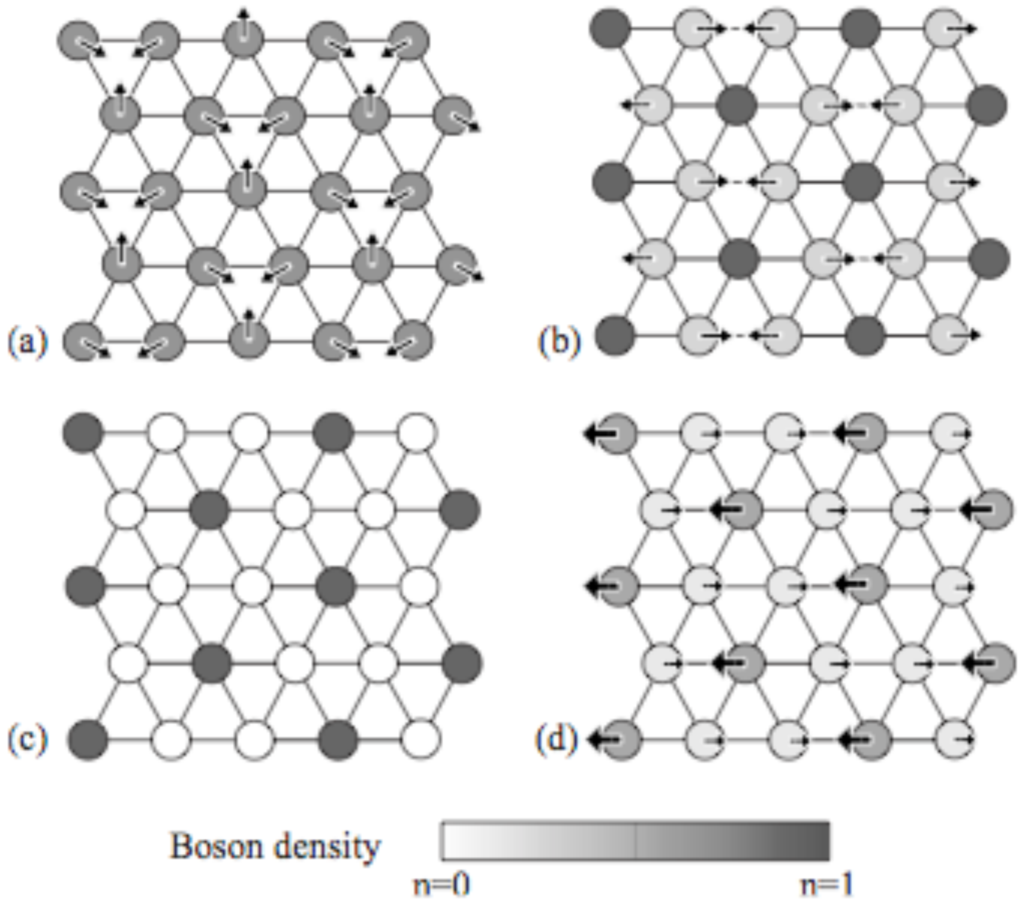
Both materials are spatially anisotropic triangular antiferromagnets

D = 2 antiferromagnets

- surprisingly complex phase diagram of spatially anisotropic triangular lattice antiferromagnet
  - no definite conclusions from numerical studies yet...
- connections with interacting boson system
  - Superfluids (XY order)
  - Mott insulators
  - Supersolids

  Andreev, Lifshitz 1969

  Nikuni, Shiba 1995
  Heidarian, Damle 2005
  Wang et al 2009
  Jiang et al 2009
  Tay, Motrunich 2010



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Triangular lattice antiferromagnet: Ising limit

PHYSICAL REVIEW VOLUME 79, NUMBER 2 JULY 15, 1950

Antiferromagnetism. The Triangular Ising Net

G. H. WANNIER
Bell Telephone Laboratories, Murray Hill, New Jersey

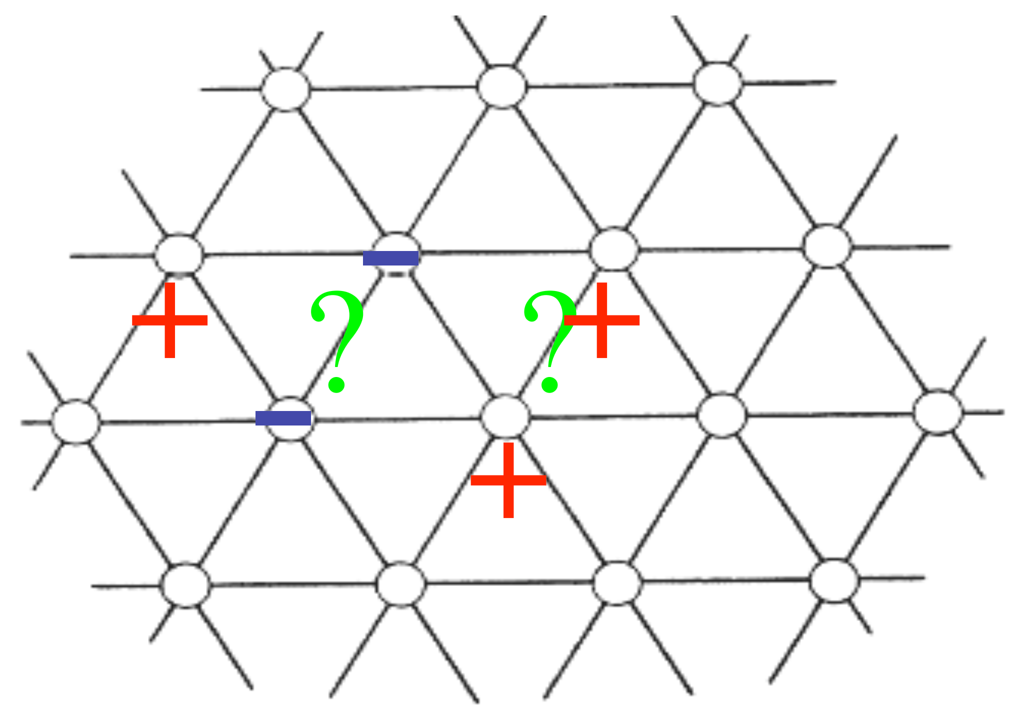
(Received February 11, 1950)

In this paper the statistical mechanics of a two-dimensionally infinite set of Ising spins is worked out for the case in which they form either a triangular or a honeycomb arrangement. Results for the honeycomb and the ferromagnetic triangular net differ little from the published ones for the square net (Curie point with logarithmically infinite specific heat). The triangular net with antiferromagnetic interaction is a sample case of antiferromagnetism in a non-fitting lattice. The binding energy comes out to be only one-third of what it is in the ferromagnetic case. The entropy at absolute zero is finite; it equals

$$S(0)=R\frac{2}{\pi}\int_0^{\pi/3}\ln(2\cos\omega)d\omega=0.3383R.$$

The system is disordered at all temperatures and possesses no Curie point.

Frustration: pairwise interactions between spins cannot be minimized simultaneously (1/3 of bonds are unhappy)



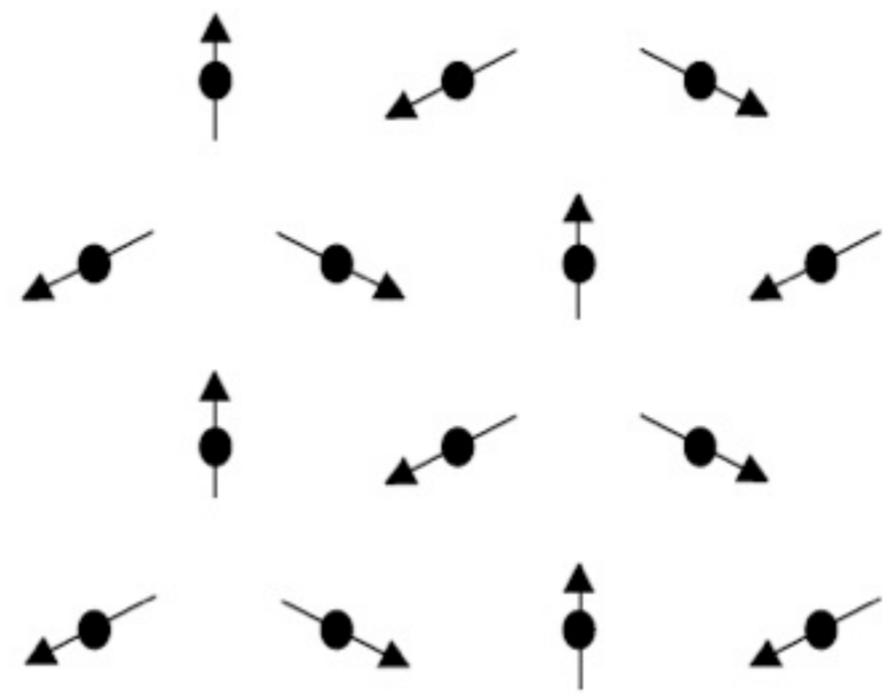
Ising spins S_r= +1 or -1 only

Heisenberg (vector) spins relieve frustration

Classical spins (unit vectors): three-sublattice 120^o structure
[commensurate spiral]

Spiral magnetic order: non-collinear but co-planar

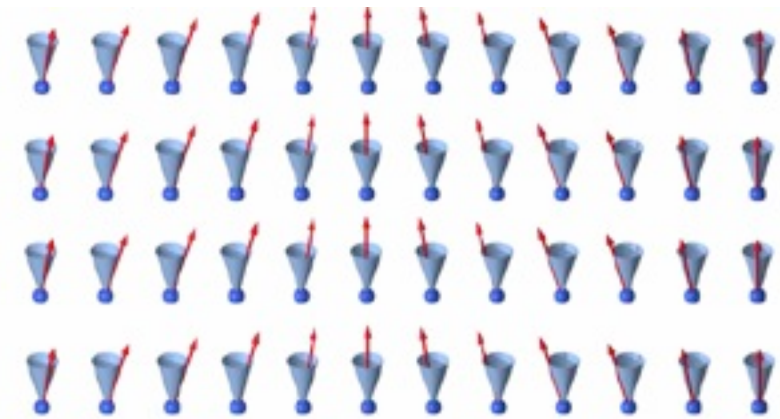
Energy per bond = $S^2 \cos(120^o) = -0.5\, S^2$



Numerical results indicate that classical 120^o structure survives down to S=1/2 limit (Singh, Huse 1992)

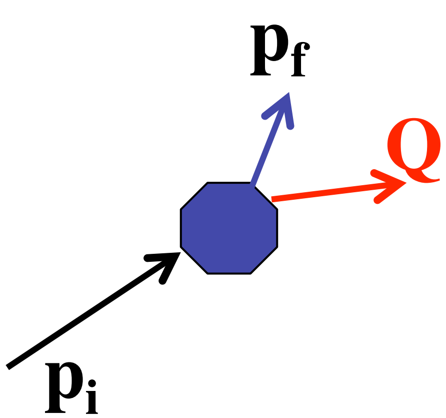
Spin waves in collinear antiferromagnet

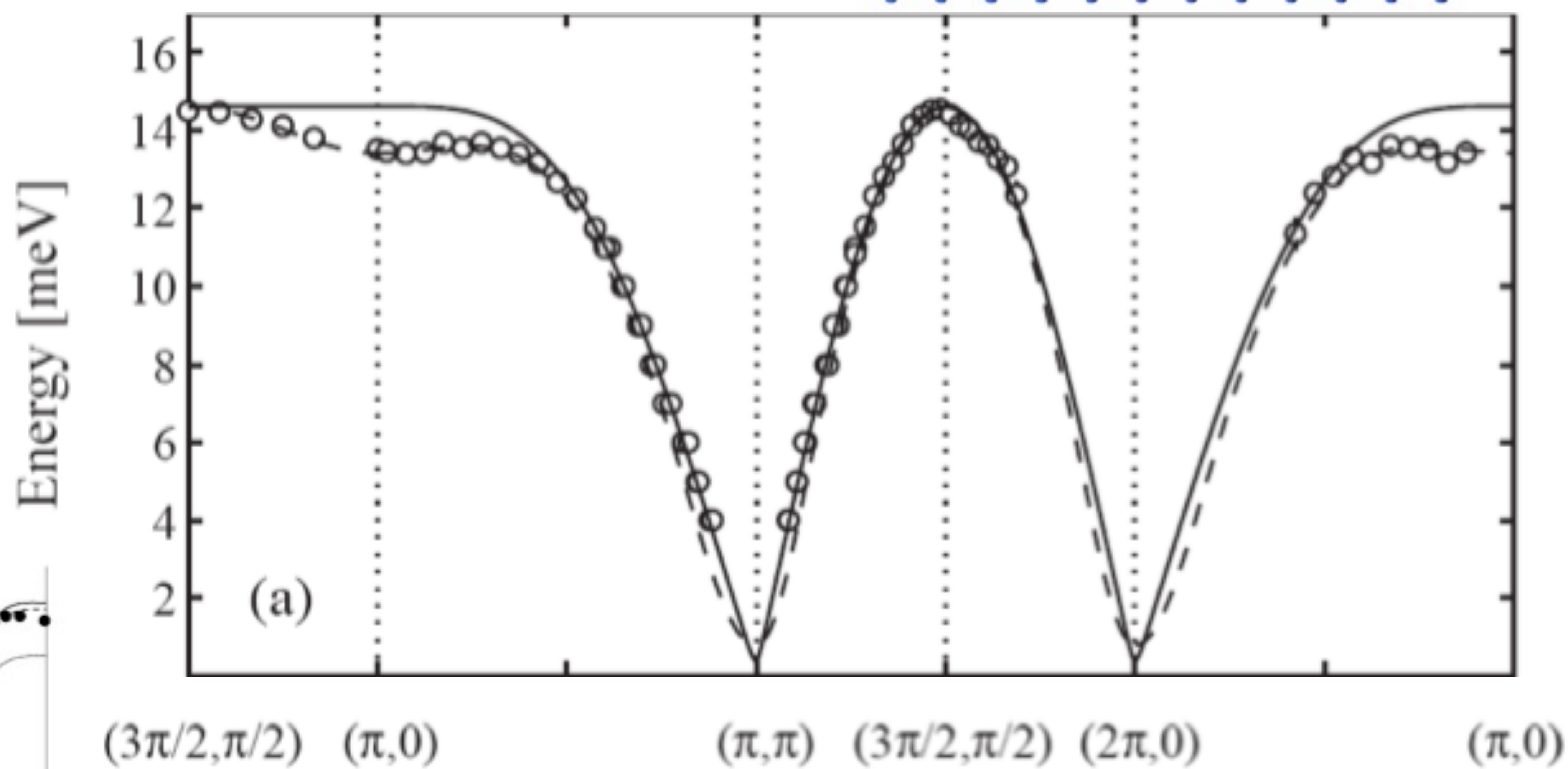
Precession of the staggered magnetization $\mathbf{N} = (-1)^r\,\mathbf{S}$

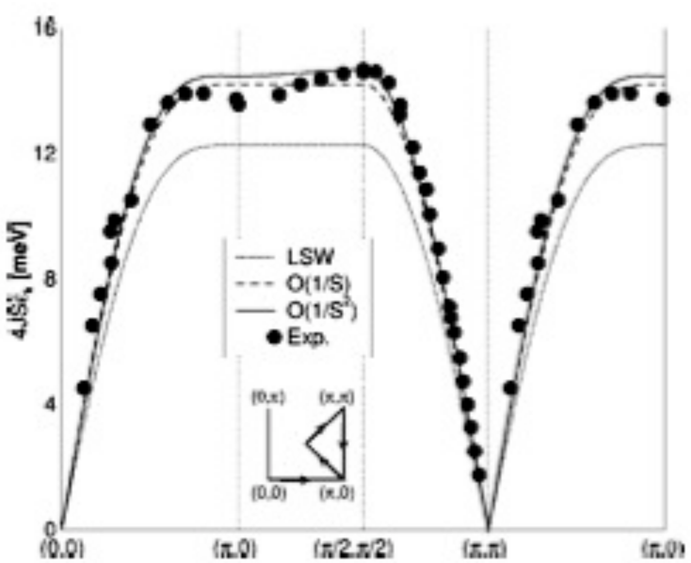


$$\left.\begin{aligned} \hbar\vec{Q} &= \vec{p}_i - \vec{p}_f \\ \omega(Q) &= \frac{\vec{p}_i^{\,2}}{2m} - \frac{\vec{p}_f^{\,2}}{2m} \end{aligned}\right\}$$

measured in inelastic neutron scattering







1/S^2: Igarashi, Nagao (2005)

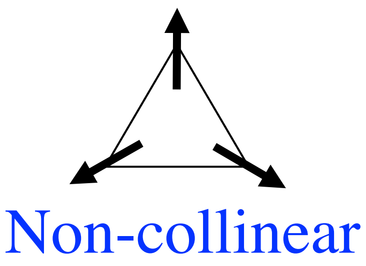
Circles: Data points for Cu(DCOO)$_2$ 4D$_2$0 (Christensen et al (2004))

Full line: Linear spin-wave theory

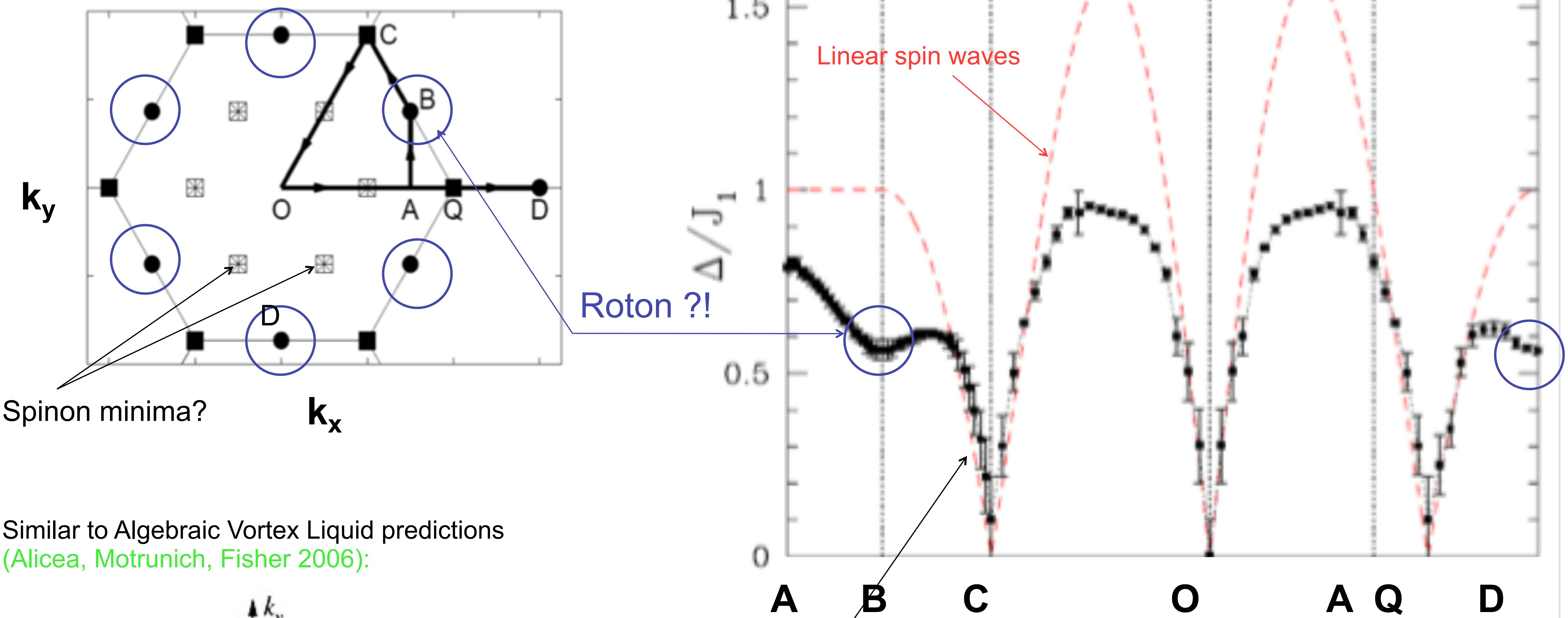
Dashed line: Series expansion result (Singh & Gelfand (1995))

Almost perfect agreement!

The same for magnons in triangular lattice?

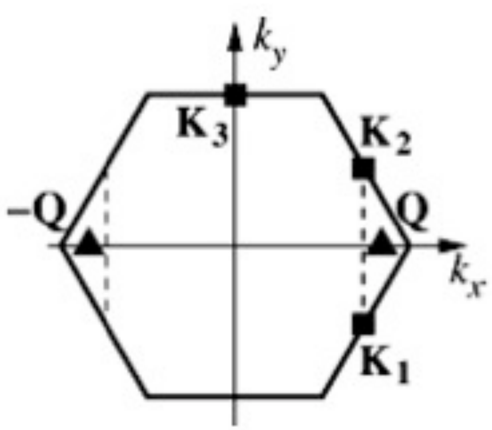


Brillouin zone:



Dots: Series expansion for S=1/2 antiferromagnet

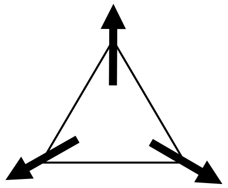
Similar to Algebraic Vortex Liquid predictions (Alicea, Motrunich, Fisher 2006):



Radical interpretation: rotons are made of spinon (fractional S=1/2 excitation) pairs.

But is this the only explanation?

PRL 96, 057201 (2006) PHYSICAL REVIEW LETTERS week ending 10 FEBRUARY 2006

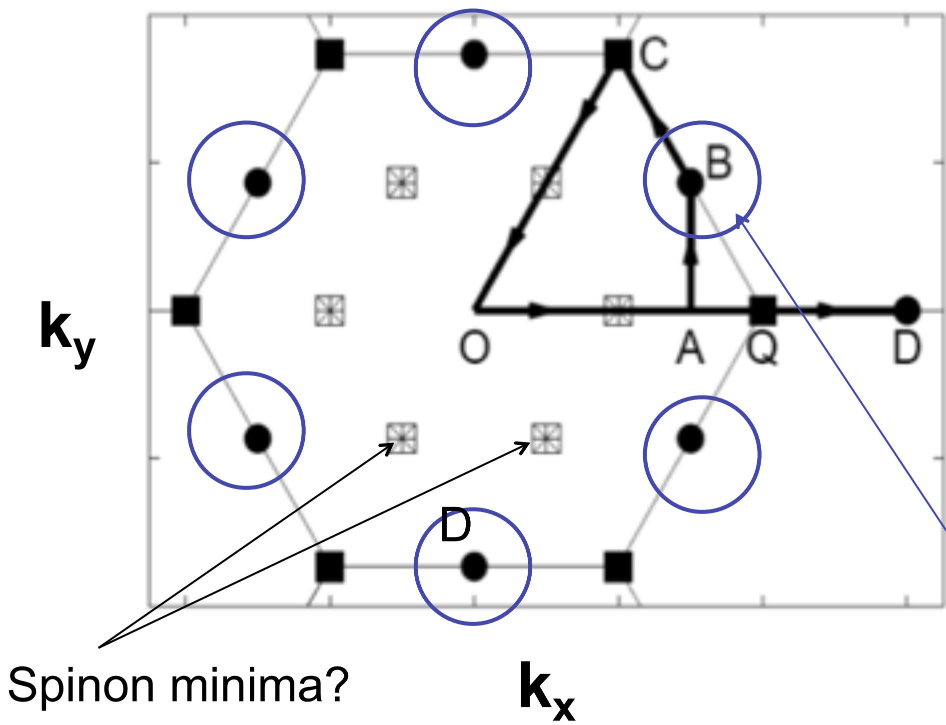


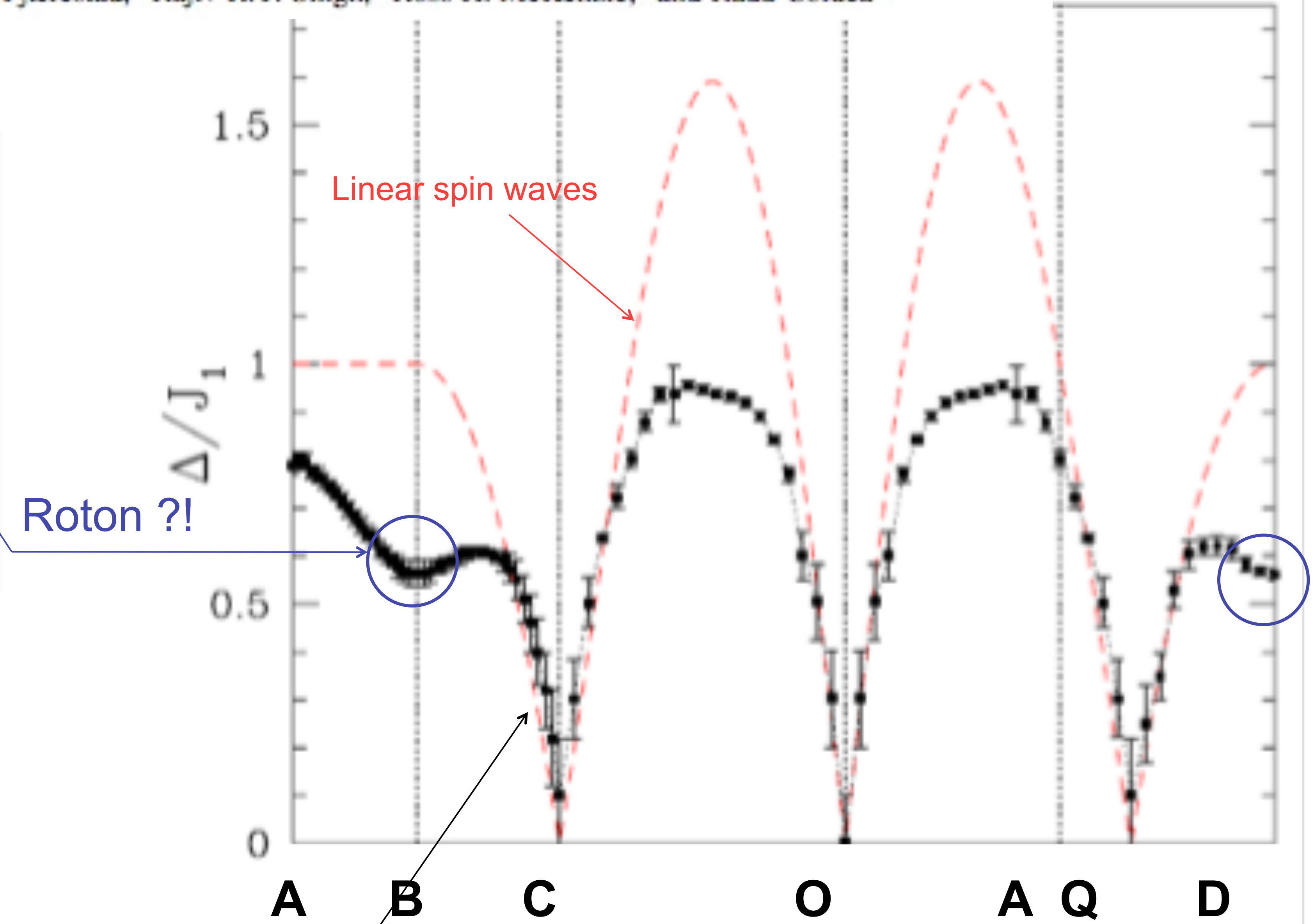
Non-collinear

Anomalous Excitation Spectra of Frustrated Quantum Antiferromagnets

Weihong Zheng,[1] John O. Fjærestad,[2] Rajiv R. P. Singh,[3] Ross H. McKenzie,[2] and Radu Coldea[4]

Brillouin zone:





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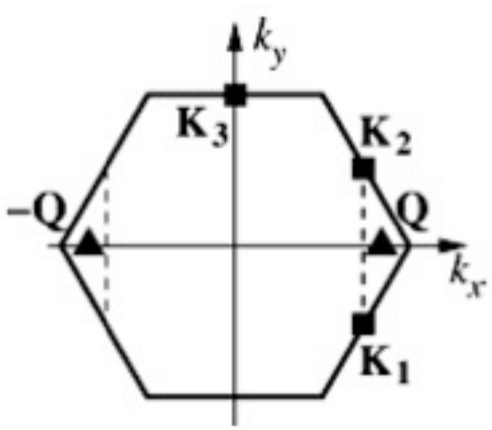

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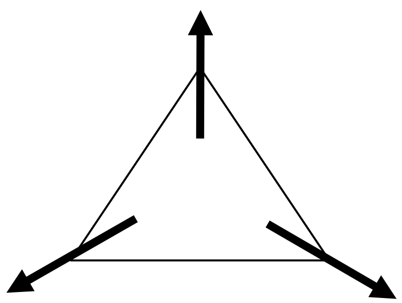
But is this the only explanation?

The minimal explanation: non-collinear spin structure is the key!



- Rotated basis: order along S^z (via rotation about S^x)

$$H = \sum_{ij} \underbrace{-\frac{1}{2}(S_i^z S_j^z + S_i^y S_j^y) + S_i^x S_j^x}_{H_{coll}} + \underbrace{\sin[\phi_i - \phi_j](S_i^z S_j^y - S_i^y S_j^z)}_{H_{non\text{-}coll}}$$

H_{coll} collinear piece: 2, 4, 6…magnons (1, S^{-1}, S^{-2} terms)

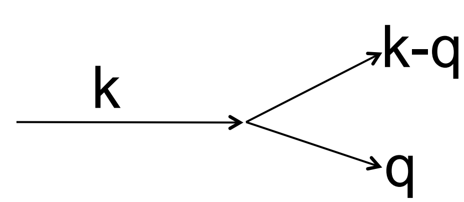
$H_{non\text{-}coll}$ non-collinear piece: 3, 5, … magnons ($S^{-1/2}$, $S^{-3/2}$ terms)

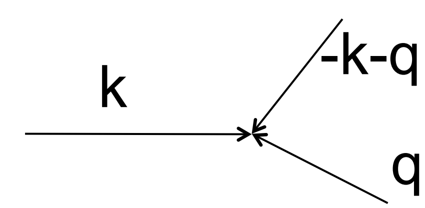
$\phi_i = \{0, 2\pi/3, 4\pi/3\}$ [angle w.r.t. fixed direction]

Spin wave expansion: S >> 1

$$S^z = S - a^\dagger a \ , \ S^x = \sqrt{\frac{S}{2}}(a^\dagger + a) \ , \ S^y = i\sqrt{\frac{S}{2}}(a^\dagger - a)$$

- $H_{non\text{-}coll}$ describes magnon decay ($a\, a^+ a^+$) and creation/annihilation ($a\, a\, a$ + h.c.)





Absent in collinear AFM (where $\phi_i = 0, \pi$)

✓ *Similar to anharmonic phonons*

- Produces 1/S (!) correction to magnon spectrum: renormalization + lifetime

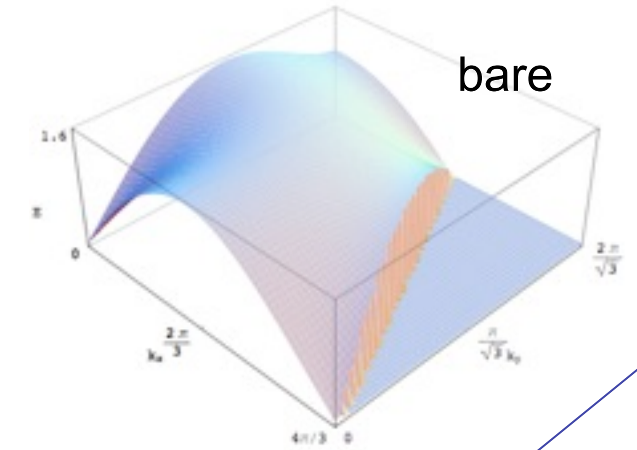
[Square lattice: corrections only at $1/S^2$ order, numerically small]

Results: 1/S corrections are HUGE

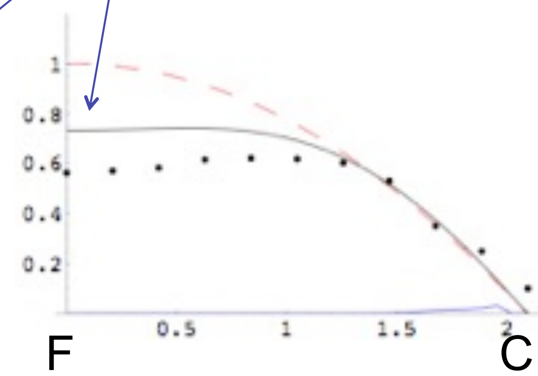
(shown in 1/4 of the Brillouin zone)

Renormalized dispersion

bare



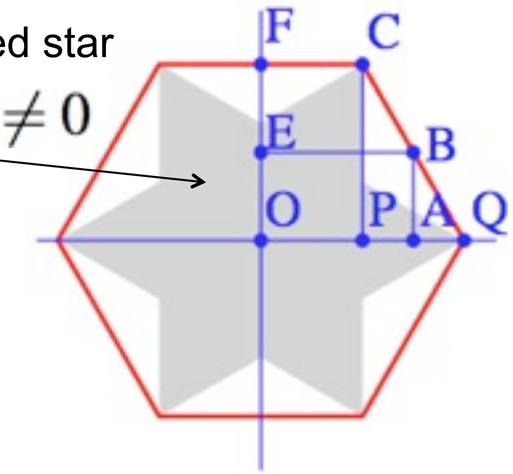
“Roton’’ minimum



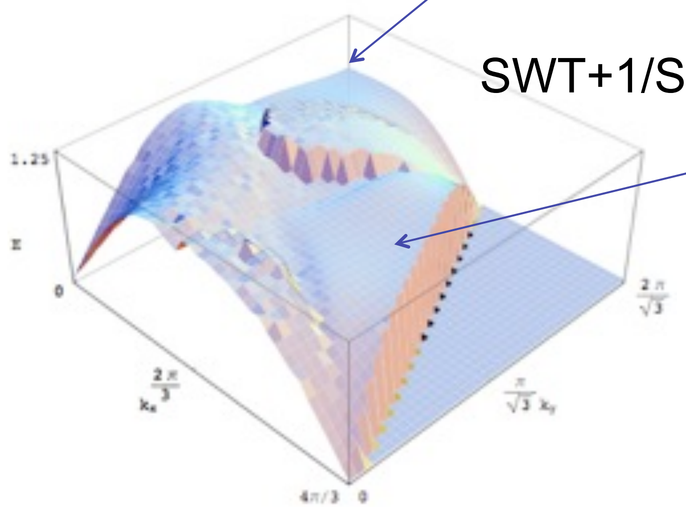
Im part (lifetime)

Shaded star

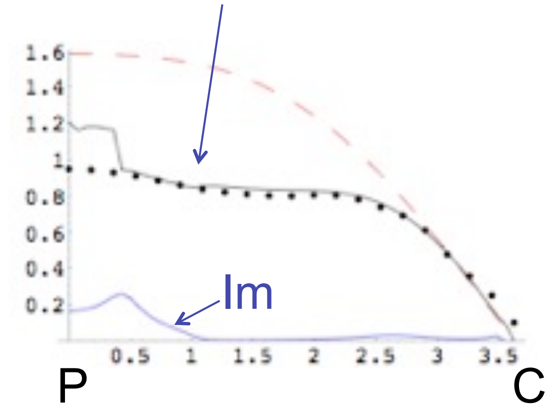
$\mathrm{Im}\Sigma \neq 0$

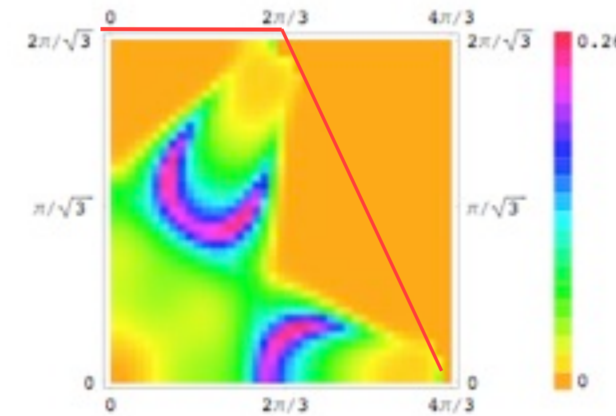


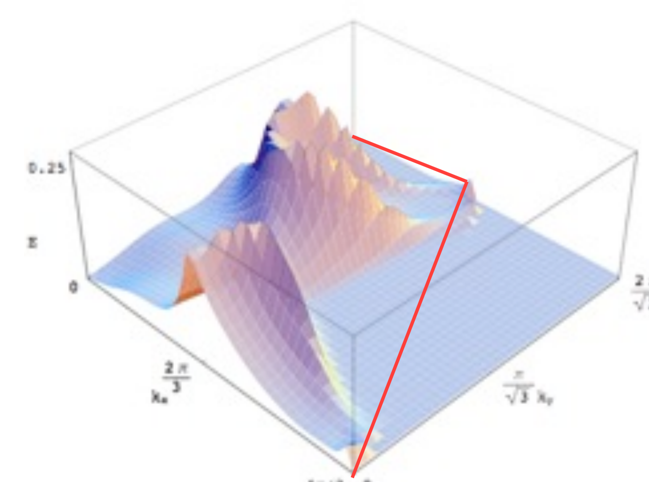
SWT+1/S



Flat dispersion





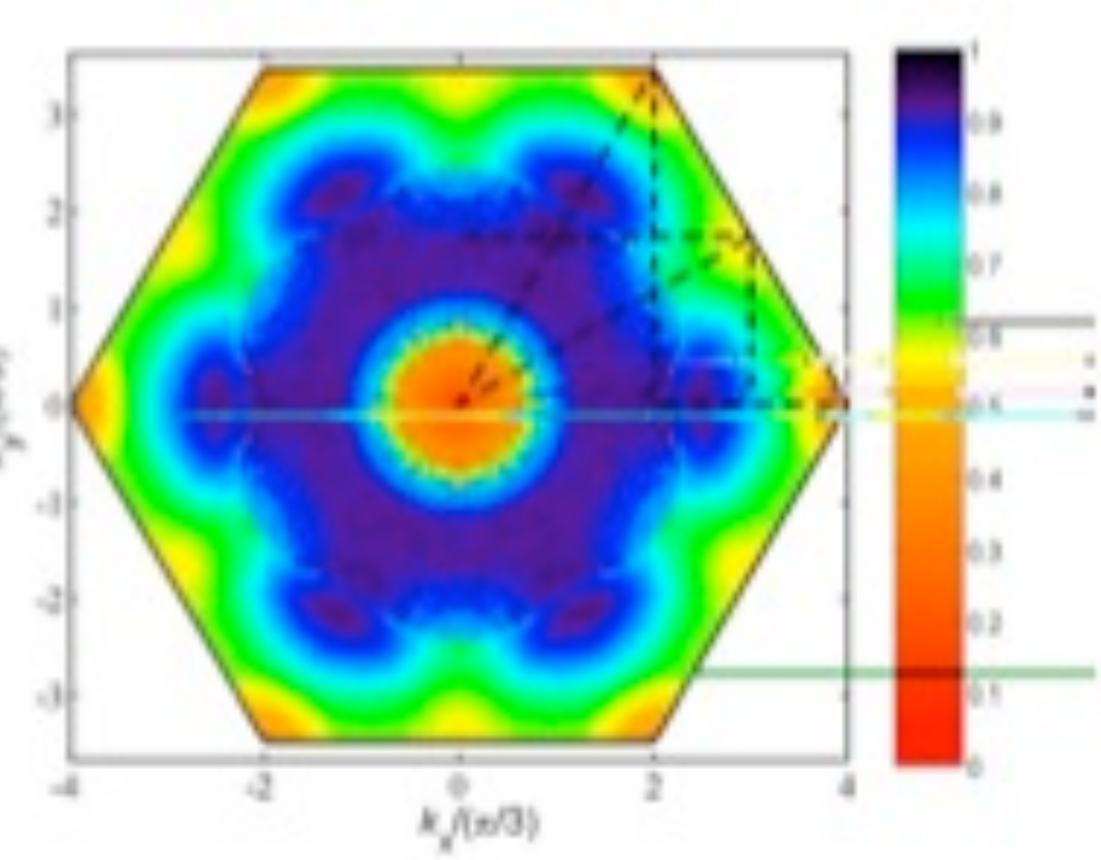


Semi-quantitative agreement with sophisticated
series expansion technique with *no adjustable parameters*
(except for S=1/2).

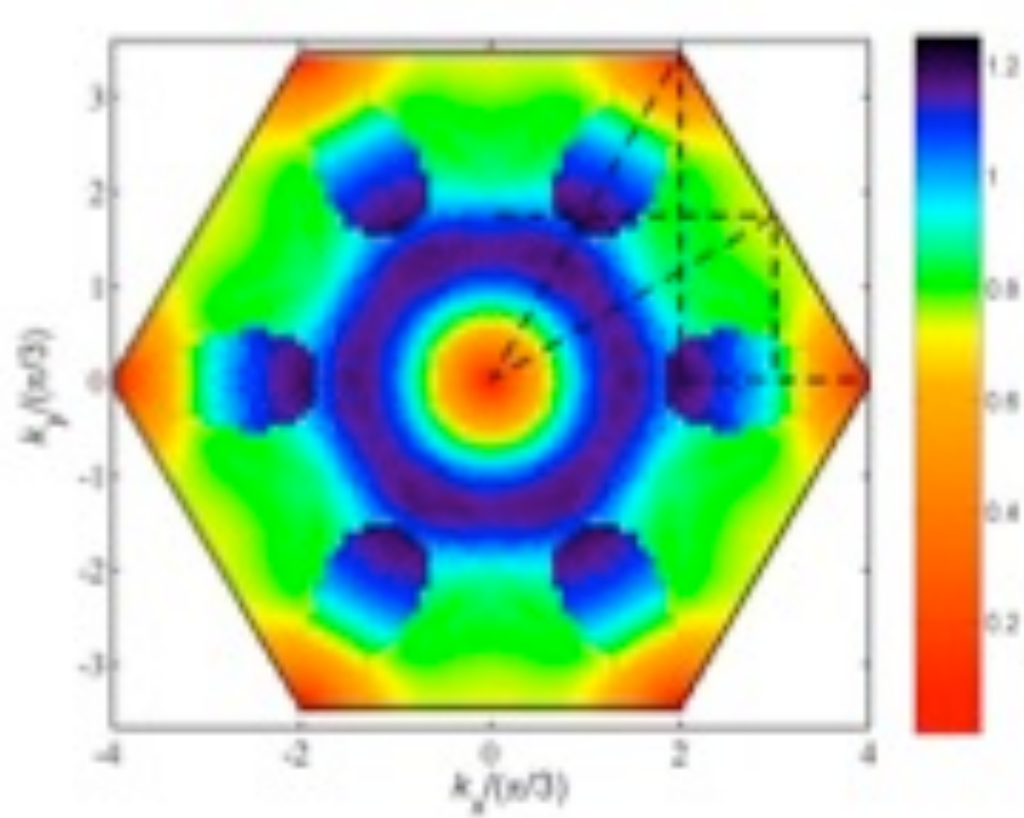
- “rotons” are part of global renormalization (weak local minimum);
- large regions of (almost) flat dispersion;
- finite lifetime [not present in numerics].

OS, Chubukov, Abanov (2006); Numerics (dots) - Zheng et al (2006); *also* Chernyshev and Zhitomirsky (2006)

Numerics



Spin waves with 1/S



Flat spin waves at high energies (but with large phase space) control thermodynamics of quantum triangular lattice antiferromagnet
down to surprisingly low temperature (of order 0.1 J) -
quite similar to He4 where rotons strongly influence finite-T state.

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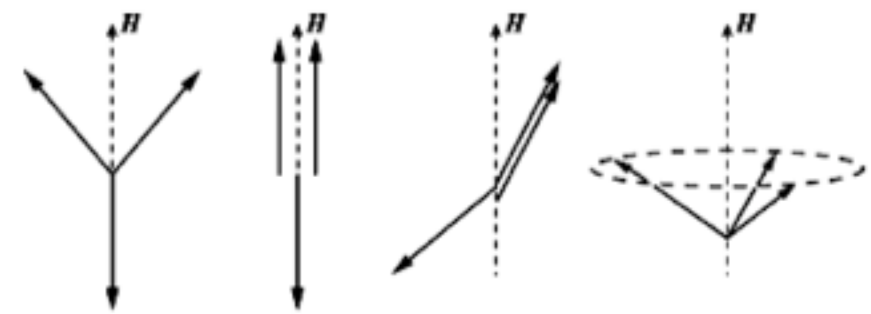
Classical isotropic Δ AFM in magnetic field

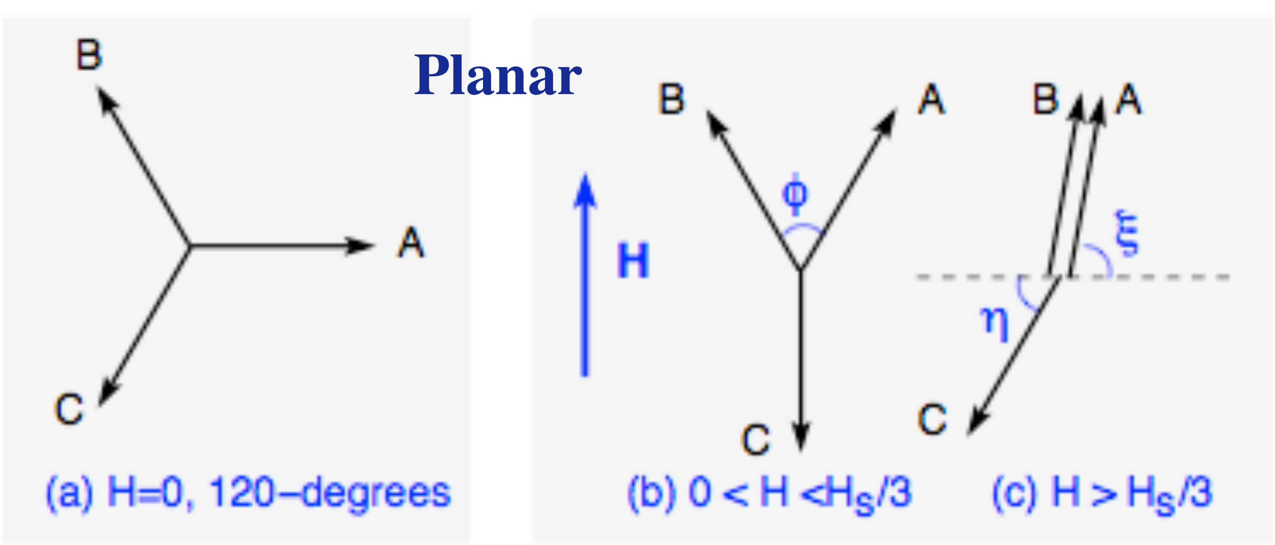


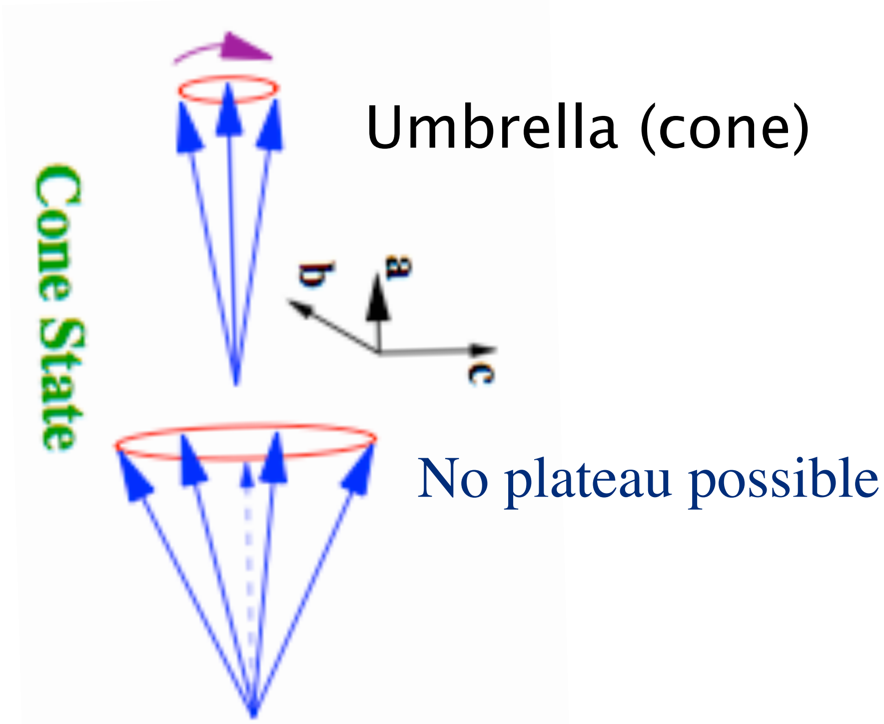
- No field: spiral (120 degree) state
- Magnetic field: **accidental degeneracy**

$$H = J\sum_{i,j}\vec{S}_i\cdot\vec{S}_j - \sum_i \vec{h}\cdot\vec{S}_i$$

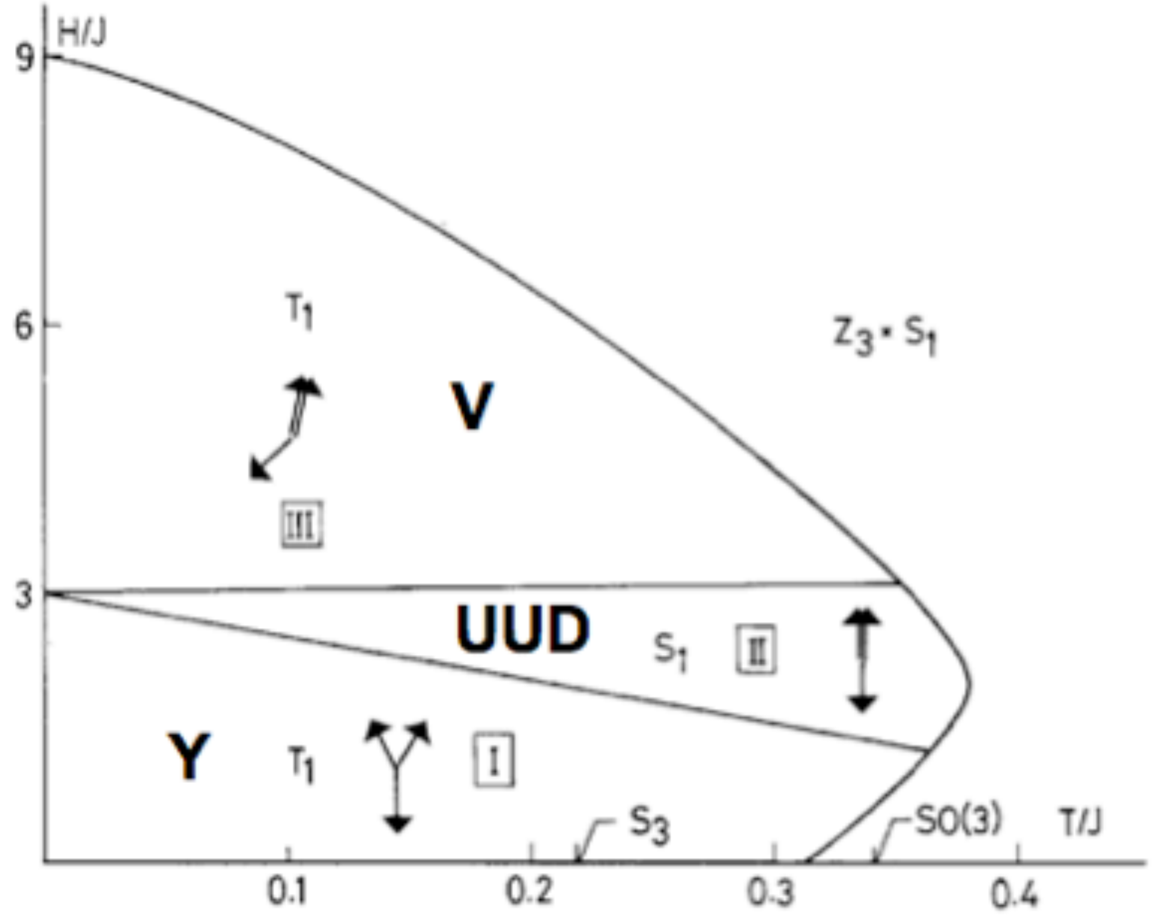
$$H = \frac{1}{2}J\sum_{\triangle}\Big(\sum_{i\in\triangle}\vec{S}_i - \frac{\vec{h}}{3J}\Big)^2$$

- all states with $\vec{S}_{i1}+\vec{S}_{i2}+\vec{S}_{i3} = \frac{\vec{h}}{3J}$ form the lowest-energy manifold
  - 6 angles, 3 equations => **2 continuous angles** (upto global U(1) rotation about **h**)

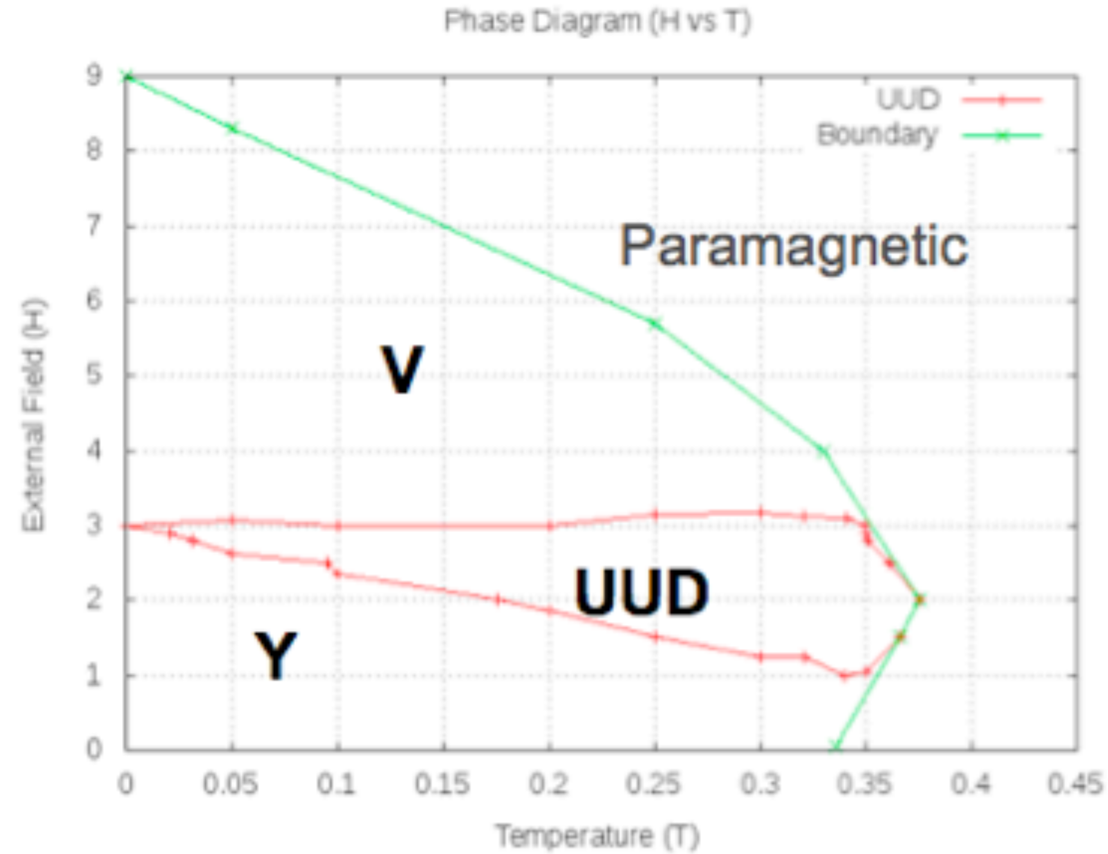




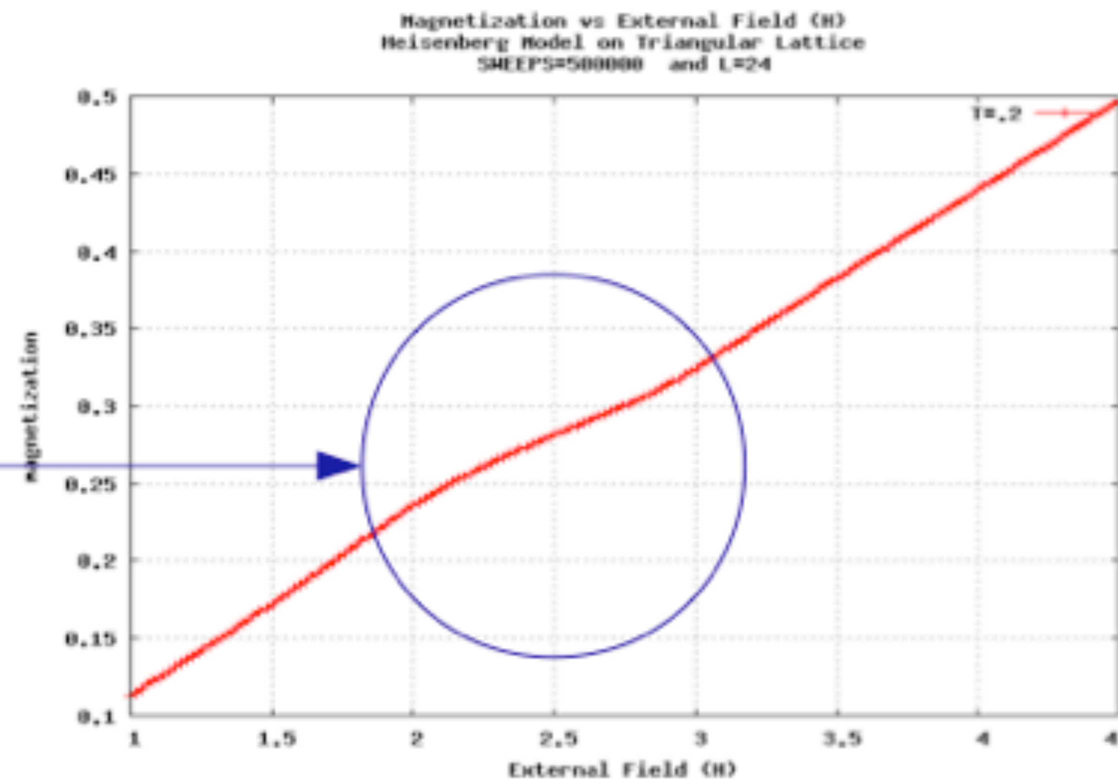
Phase diagram



H. Kawamura, S. Miyashita:
JPSJ (1985)



Head, Griset, Alicea, OS 2010



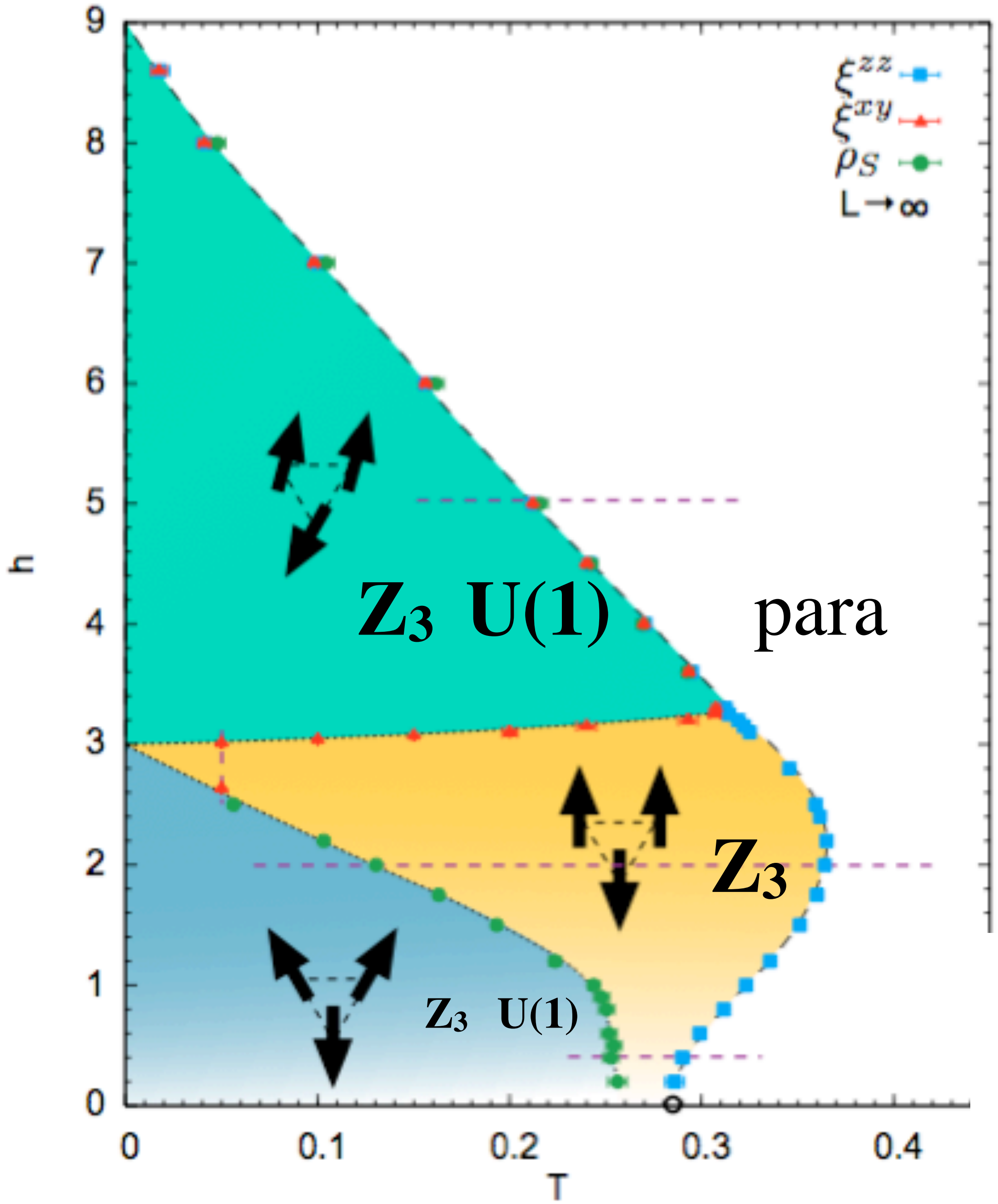
Entropic Selection:

- Planar states favored by thermal fluctuations
- UUD state around m=1/3 resulting in quasi-plateau

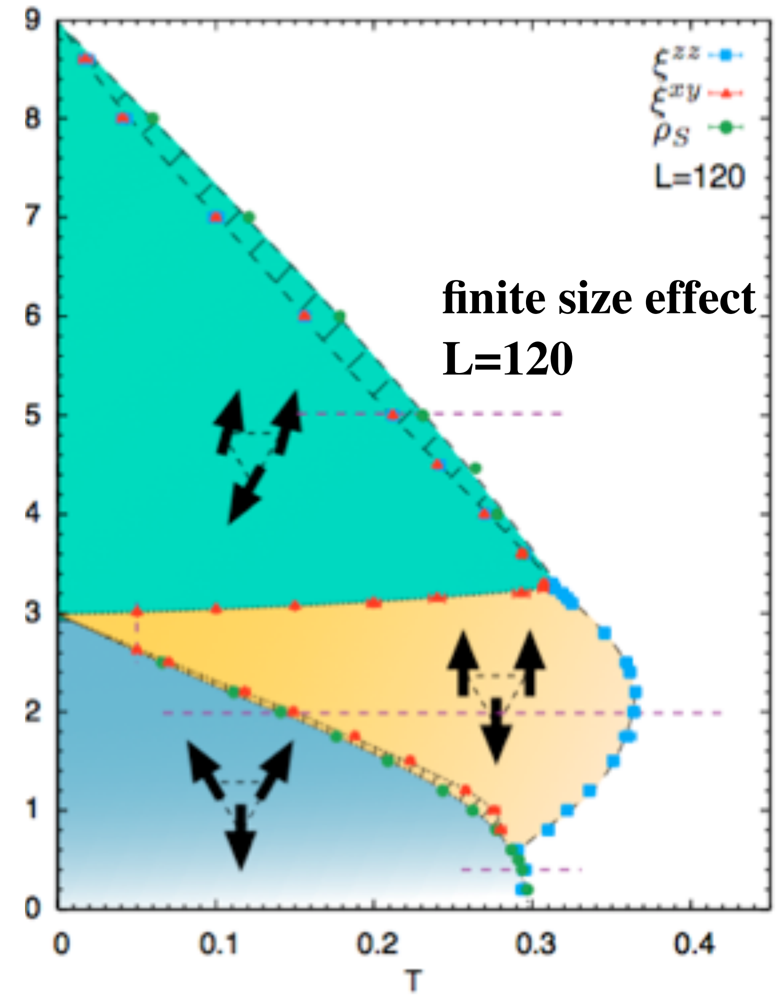
Finite T: minimize F = E - T S

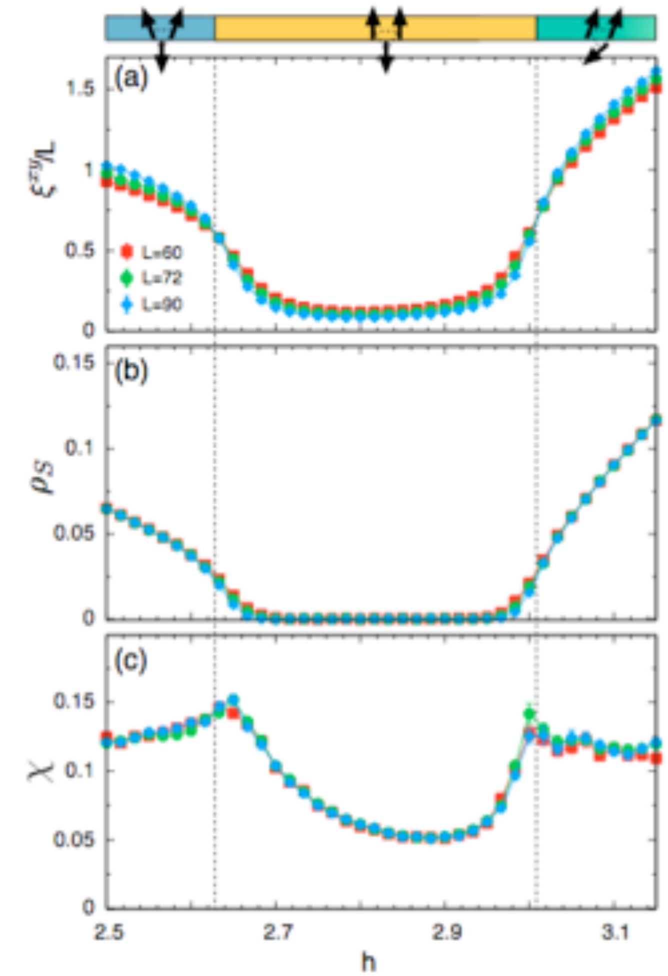
Planar states have higher entropy!

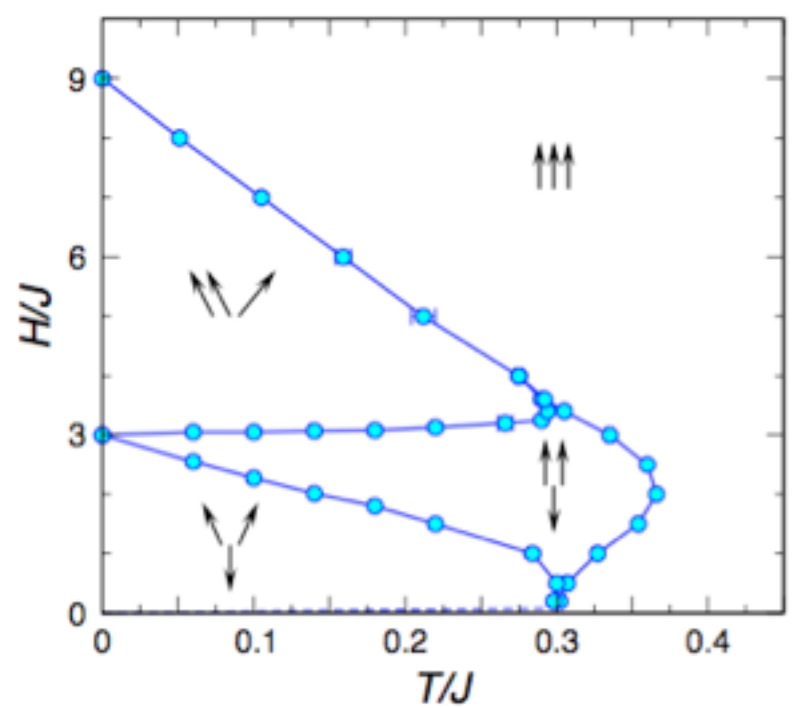
Phase diagram of the classical model: Monte Carlo



Seabra, Momoi, Sindzingre, Shannon 2011





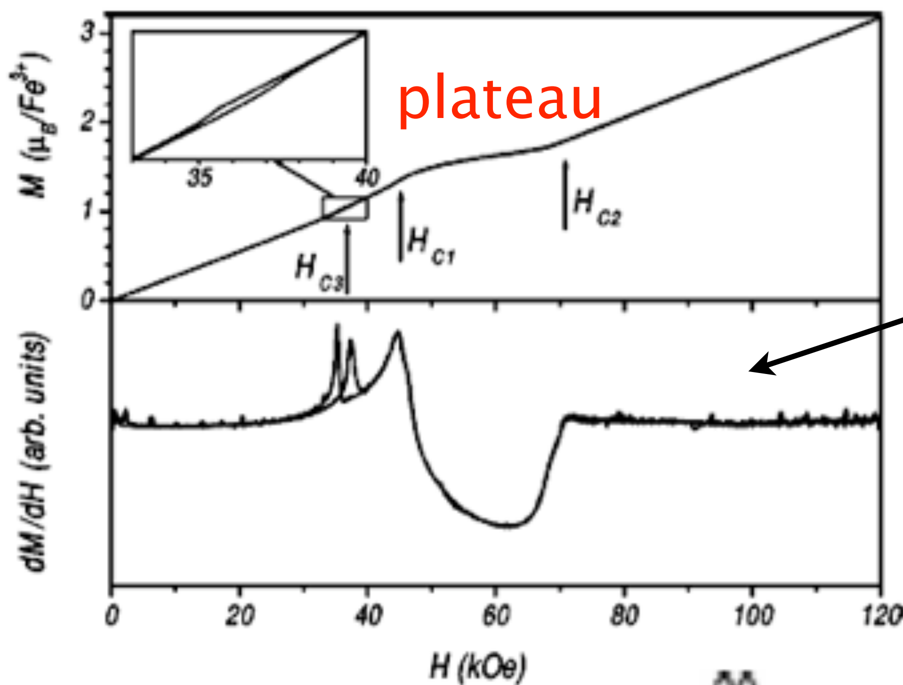


Gvozdikova, Melchy, Zhitomirsky 2010

Experimental realizations

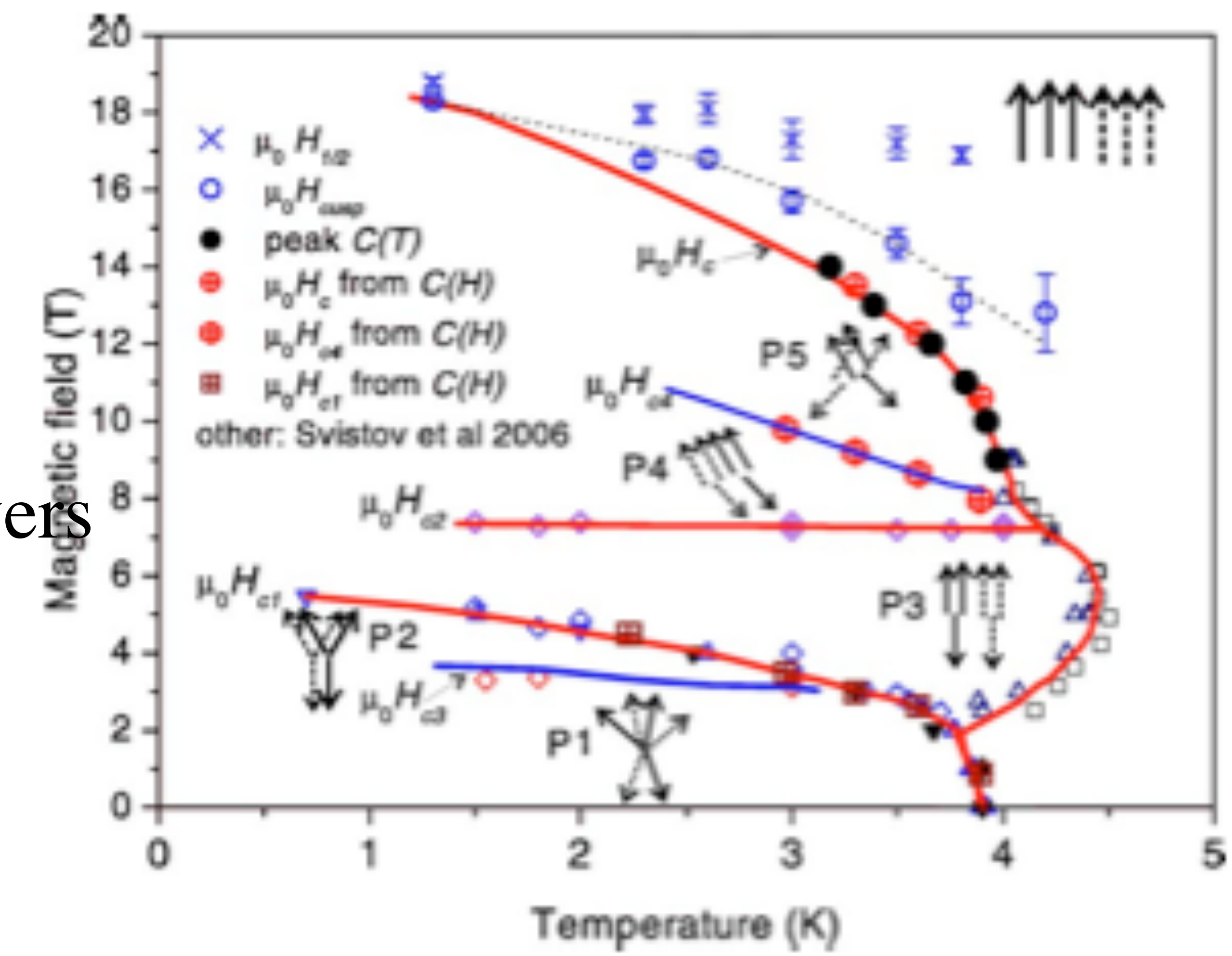
RbFe(MoO_4)$_2$:
S=5/2 Fe^{3+}

Svistov et al PRB (2003)
Smirnov et al PRB (2007)



plateau is signaled by depression in dM/dH

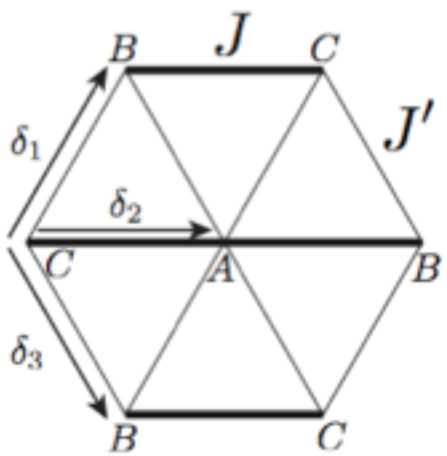
Phase diagram contains only co-planar states

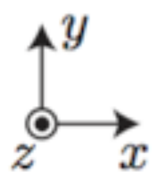


Two antiferromagnetically coupled layers

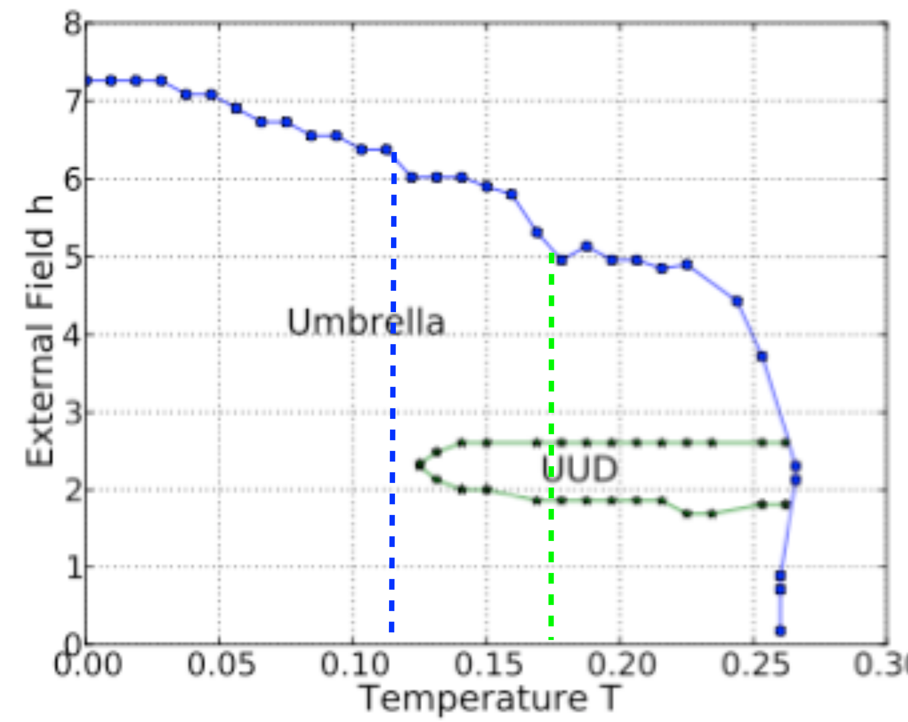
Plateau width increases with T

Effect of spatial anisotropy J” < J: energy vs entropy

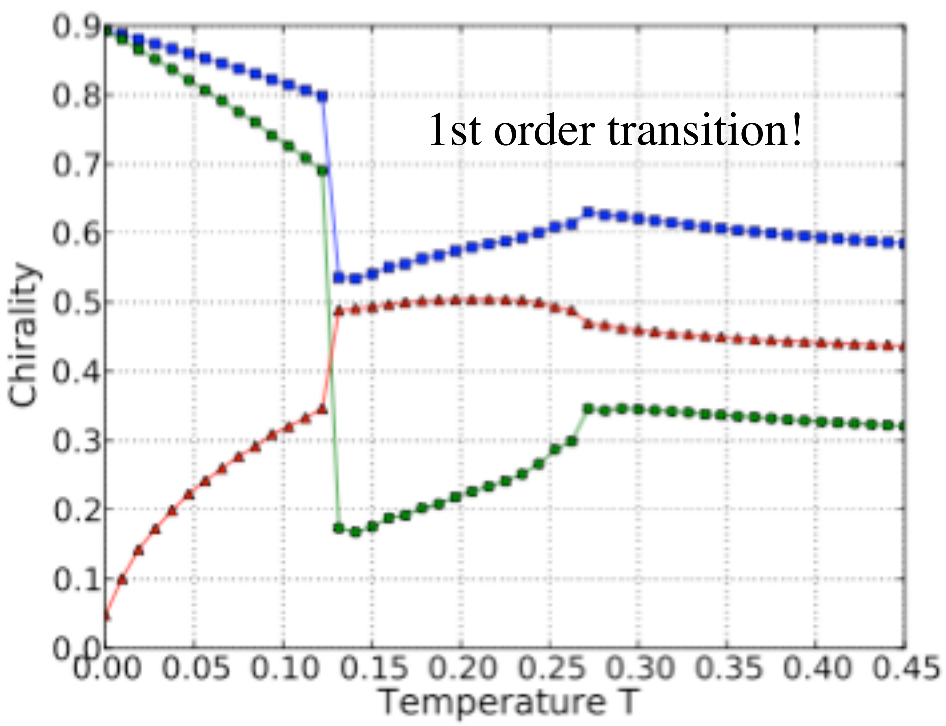


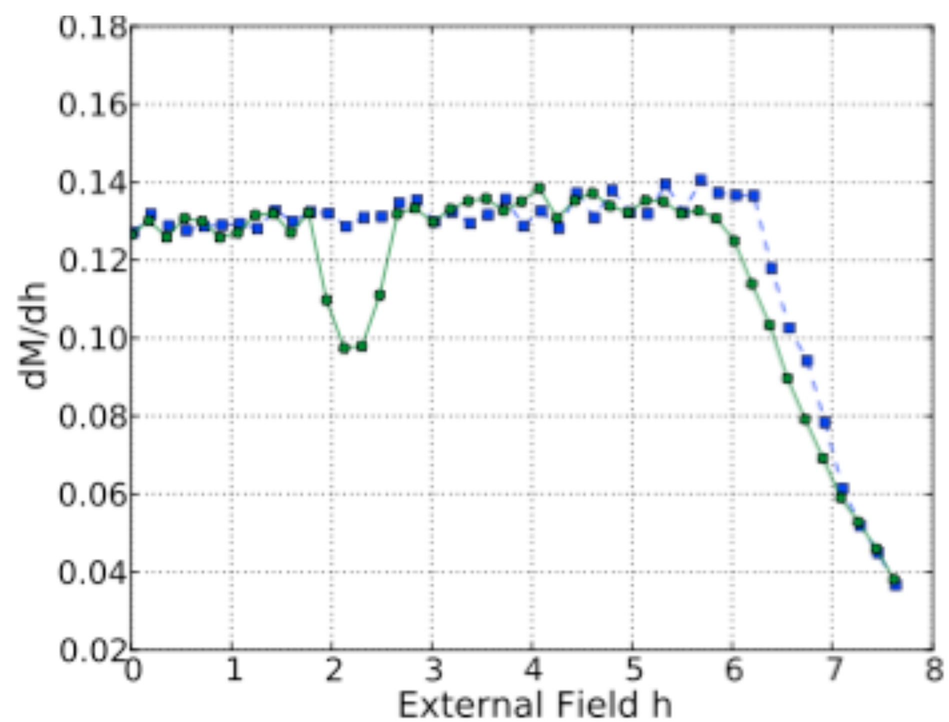


Umbrella state:
favored classically,
energy gain (J-J’)^2/J



J’ = 0.765 J





$$\boldsymbol{\kappa} = \frac{2}{3\sqrt{3}}\frac{1}{N}\sum_{\mathbf{r}}\Big(\mathbf{S}_{\mathbf{r}} \times \mathbf{S}_{\mathbf{r}+\boldsymbol{\delta}_1} + \\ + \mathbf{S}_{\mathbf{r}+\boldsymbol{\delta}_1} \times \mathbf{S}_{\mathbf{r}+\boldsymbol{\delta}_2} + \mathbf{S}_{\mathbf{r}+\boldsymbol{\delta}_2} \times \mathbf{S}_{\mathbf{r}}\Big)$$

Low T: energetically preferred umbrella
High T: entropically preferred UUD
Y and V are less stable.

Outline

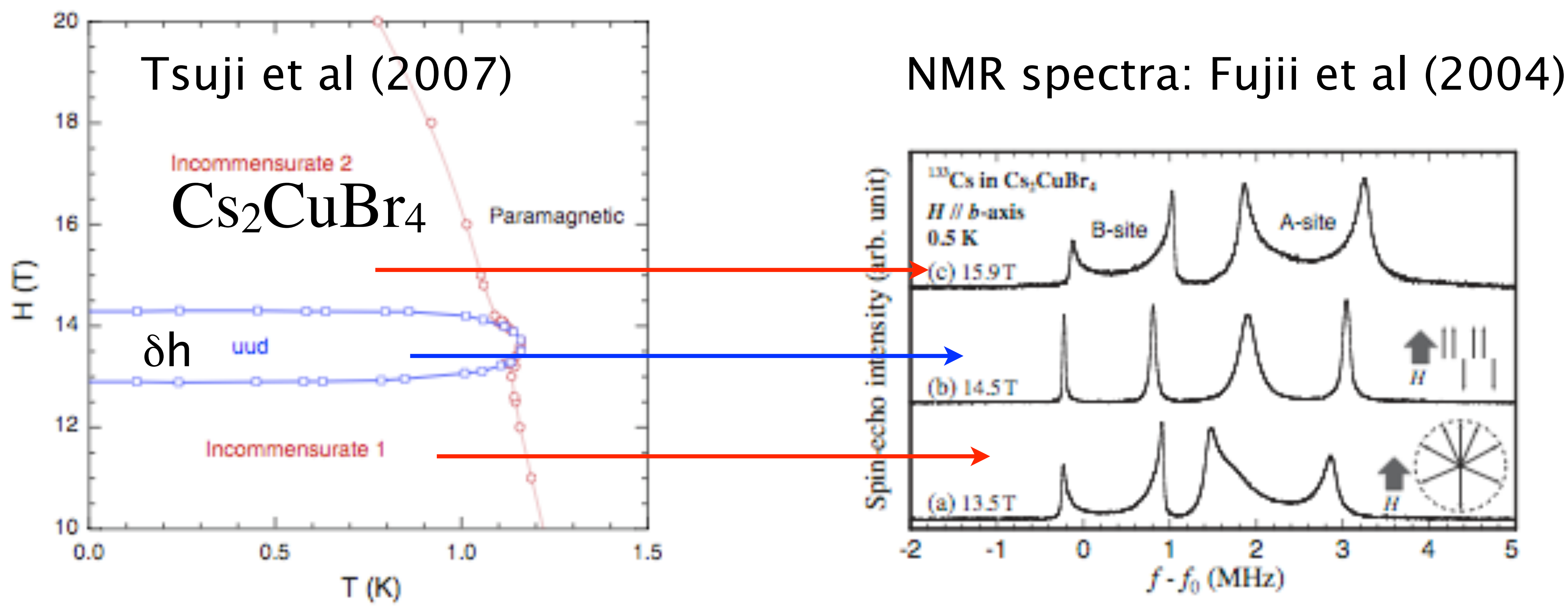
- motivation: Cs_2CuBr_4 , Cs_2CuCs_4
- Spin waves in non-collinear spin structures
- classical antiferromagnet in a field: entropic selection
  - spatial anisotropy - high-T stabilization of the plateau
- Quantum spins: zero-point fluctuations
  - Large-S analysis of interacting spin waves
- Approach from one dimension
  - sequence of plateaux and selection rules

★ (attempt at) Unification

Order-from-Disorder via Quantum fluctuations: finite S effect

- fluctuation spectra of *different* spin structures are *different*: $E = E_{class} + \Delta E^{sw}$
- quantum fluctuations prefer planar arrangement $\Delta E^{sw}_{planar} = \frac{S}{2}\sum_k \omega_{planar}(k) < \Delta E^{sw}_{umbrella} = \frac{S}{2}\sum_k \omega_{umbrella}(k)$
- prefer collinear configuration even more, when possible: state with maximum number of soft modes wins
- plateau is a quantum effect, width $\delta h = 1.8\, J/(2S)$ ($h_{saturation} = 9\, J$)

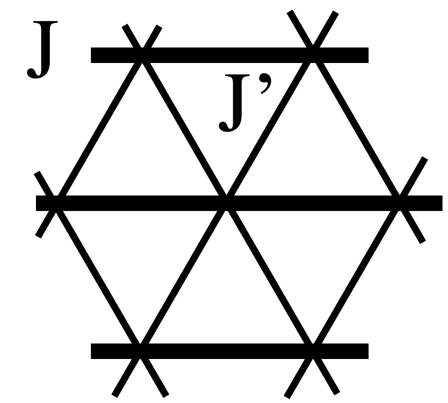
  Chubukov, Golosov (1991)
- plateau is the effect of interactions (hence, width ~ 1/S) between spin waves



plateau: T-independent width, quantum effect

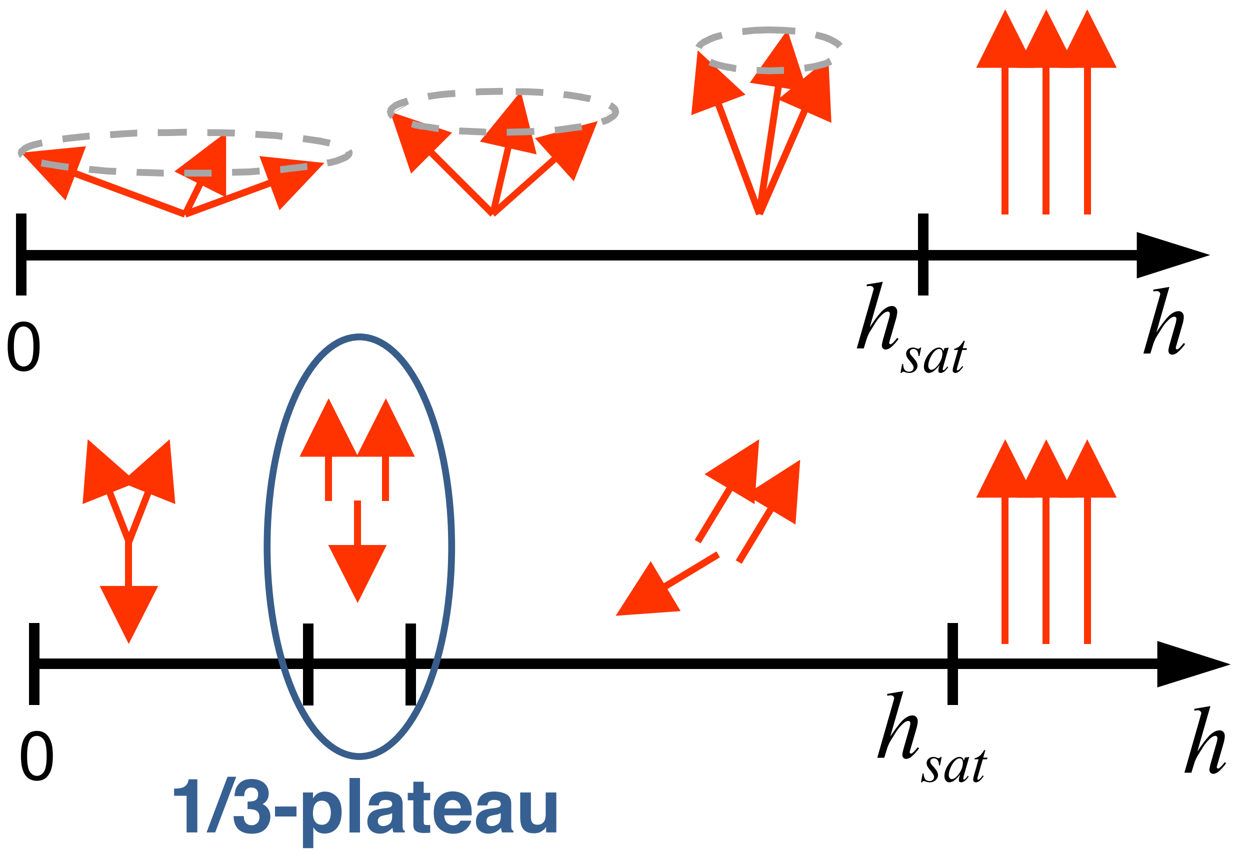
Spatially anisotropic model: classical prediction *fails*

$$H = \sum_{\langle ij \rangle} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j - h \sum_i \mathbf{S}_i^z$$



T=0

$J' \neq J$



Umbrella state: favored classically; energy gain $(J\text{-}J')^2/J$

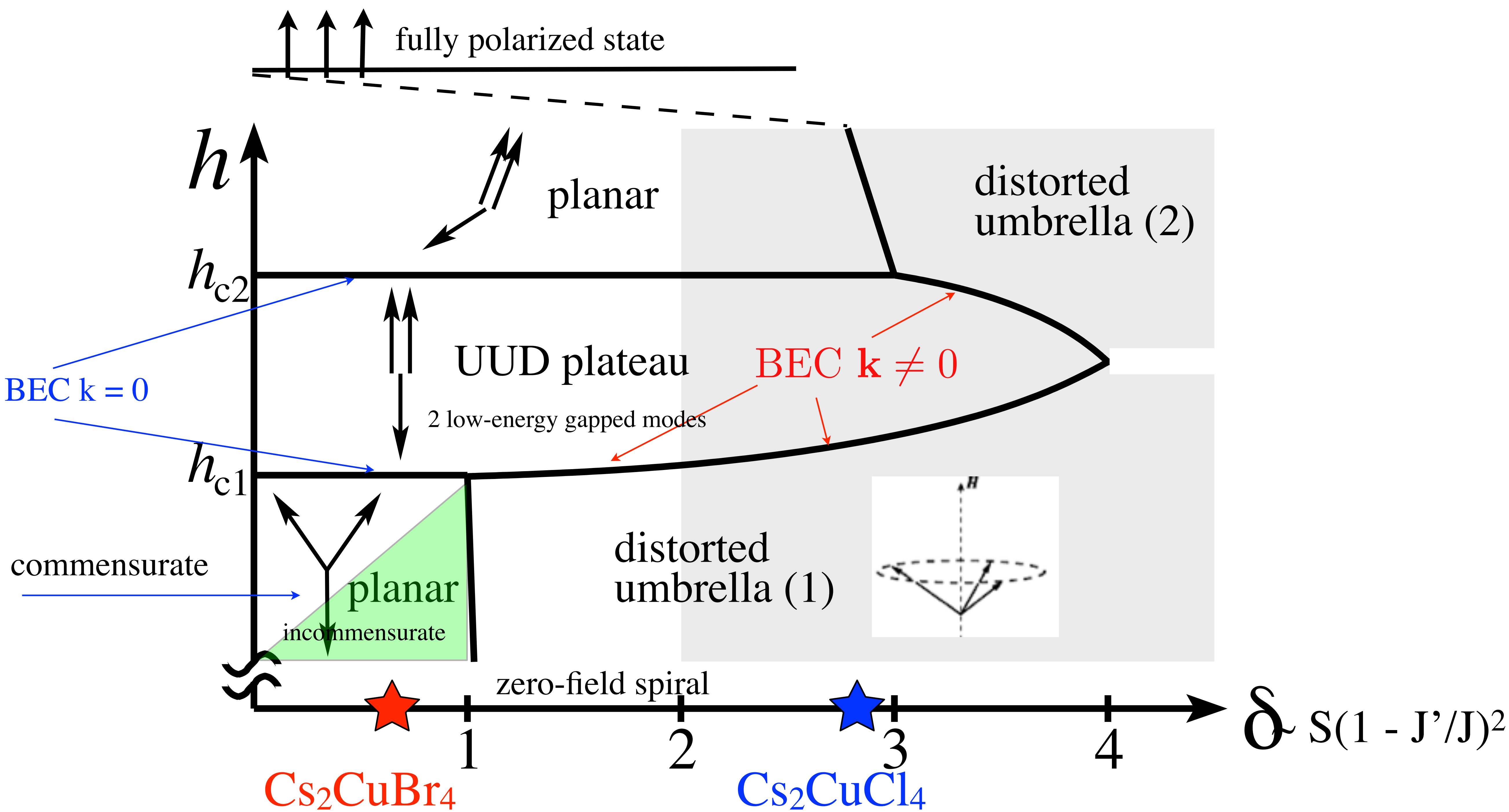
Planar states: favored by quantum fluctuations; energy gain J/S

The competition is controlled by dimensionless parameter

$$\delta = S(J - J')^2/J^2$$

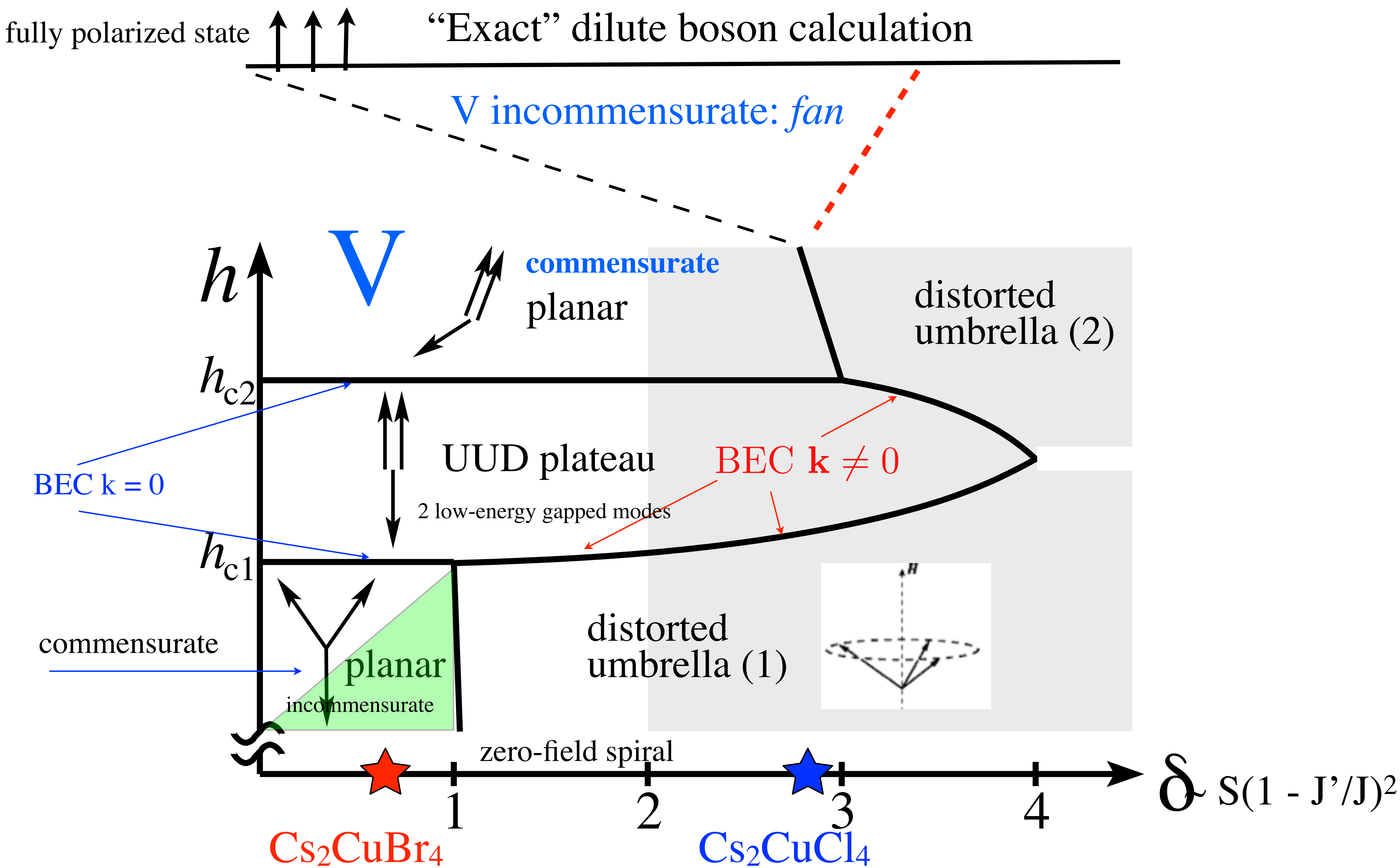
Our *semiclassical* approach: treat spatial anisotropy (J–J') as a perturbation to **interacting** spin waves

- single dimensionless parameter $\delta = S(1 - J'/J)^2$:



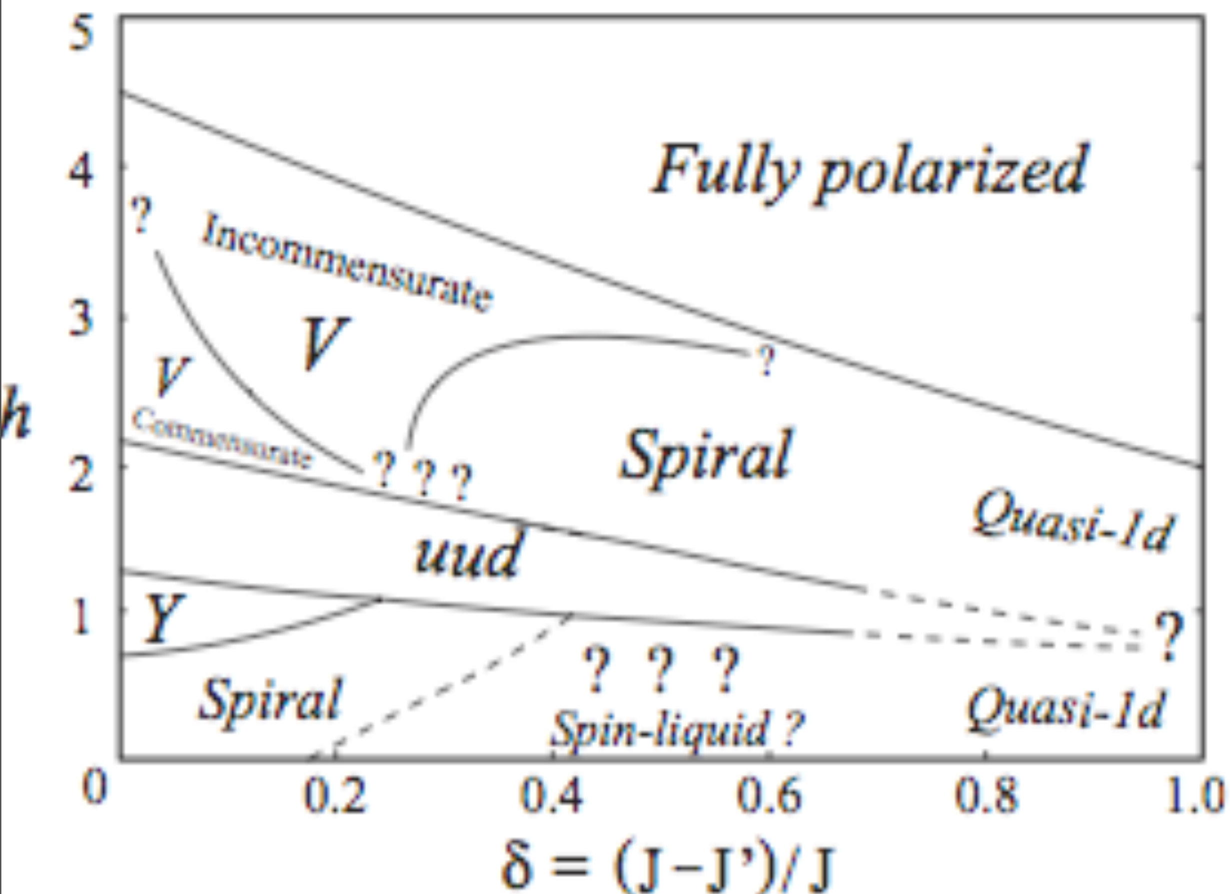
Alicea, Chubukov, Starykh PRL 102, 137201 (2009)

More detailed phase diagram

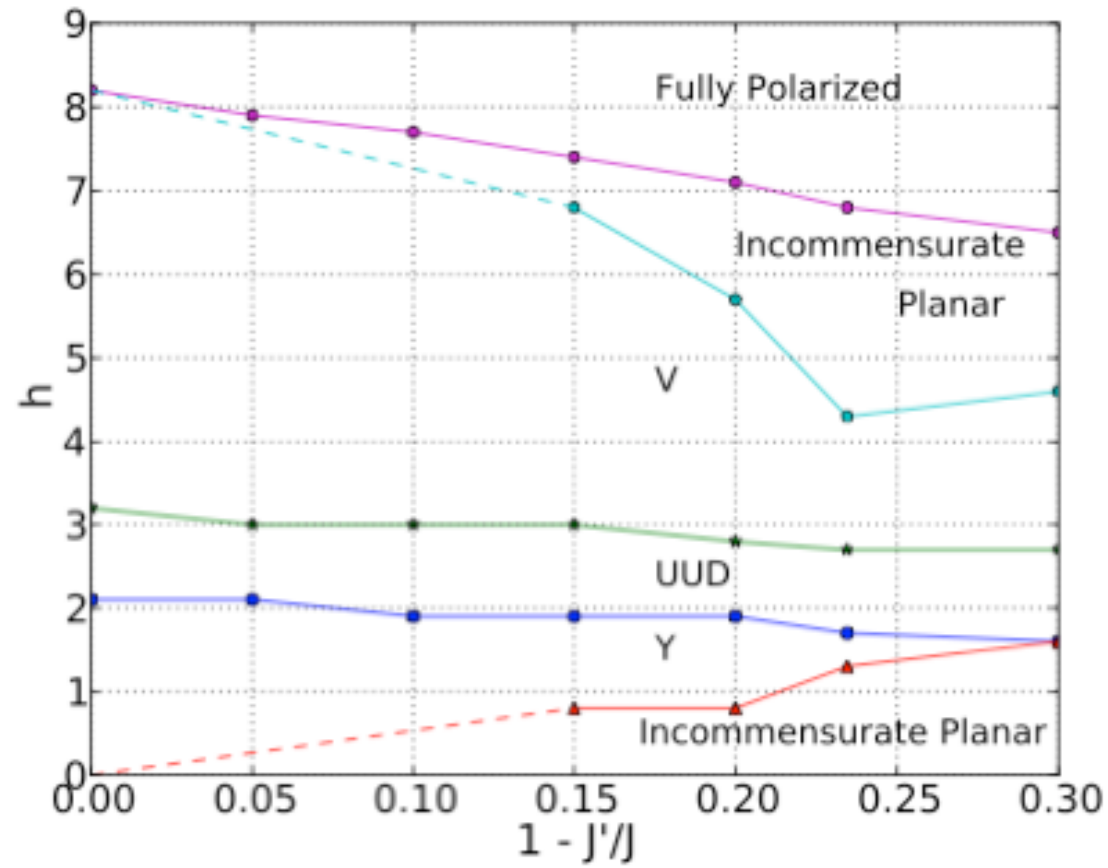


Alicea, Chubukov, Starykh PRL 102, 137201 (2009)

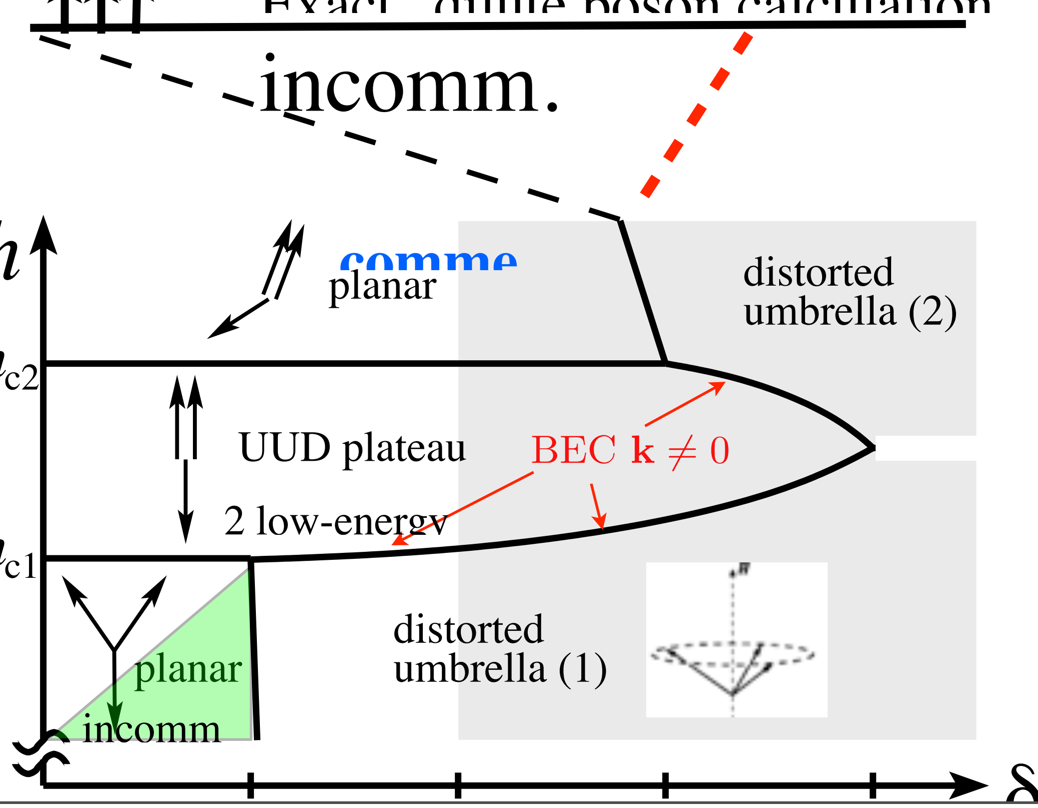
Variational wave function calculation
Tay, Motrunich 2010



Modeling quantum spins by **biquadratic** interaction
Griset, Head, Alicea, OS (2011)



“Exact” dilute boson calculation



Main effect of J’/J < 1:
✦ appearance of *incommensurate* states but spin configurations remain *co-planar*
✦ magnetization plateau is insensitive to spatial anisotropy J’/J

Outline

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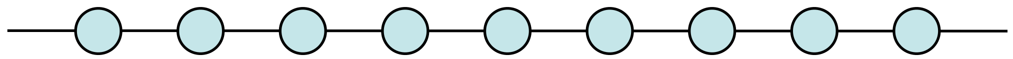
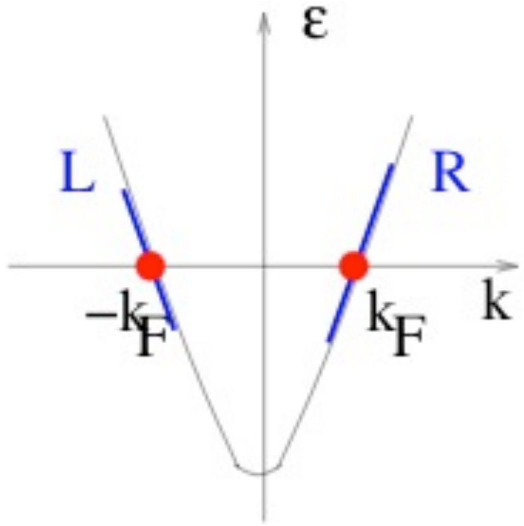
★ (attempt at) Unification

Heisenberg spin chain via free Dirac fermions

- Spin-1/2 AFM chain = half-filled (1 electron per site, $k_F=\pi/2a$) fermion chain



- Spin-charge separation



$$H_{\text{dirac}} = iv \int dx \sum_{s=\uparrow,\downarrow} (\Psi^+_{L,s}\partial_x \Psi_{L,s} - \Psi^+_{R,s}\partial_x \Psi_{R,s})$$

- *q=0 fluctuations: right- and left- spin currents*

$$\vec{J}_R = \Psi^+_{Rs} \frac{\vec{\sigma}_{ss'}}{2} \Psi_{Rs'} \;,\; \vec{J}_L = \Psi^+_{Ls} \frac{\vec{\sigma}_{ss'}}{2} \Psi_{Ls'}$$

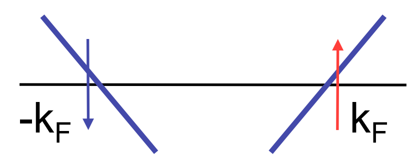
- *$2k_F$ (= π/a) fluctuations: charge density wave ε , spin density wave N*

Susceptibility

Staggered Magnetization N

$$\begin{cases} N^+ \sim \Psi^+_{R\uparrow}\Psi_{L\downarrow} + \text{h.c.} \\ N^z \sim \Psi^+_{R\uparrow}\Psi_{L\uparrow} - \Psi^+_{R\downarrow}\Psi_{L\downarrow} + \text{h.c.} \end{cases}$$

Spin flip ΔS=1



1/q

$\chi_{1d}(q)$

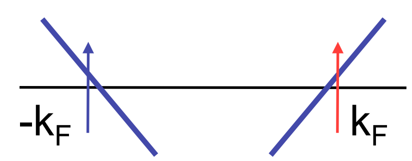
1/q

Staggered Dimerization

ε = $(-1)^x S_x S_{x+a}$

$$\varepsilon \sim i\left(\Psi^\dagger_{R\uparrow}\Psi_{L\uparrow} + \Psi^\dagger_{R\downarrow}\Psi_{L\downarrow} - \text{h.c.}\right)$$

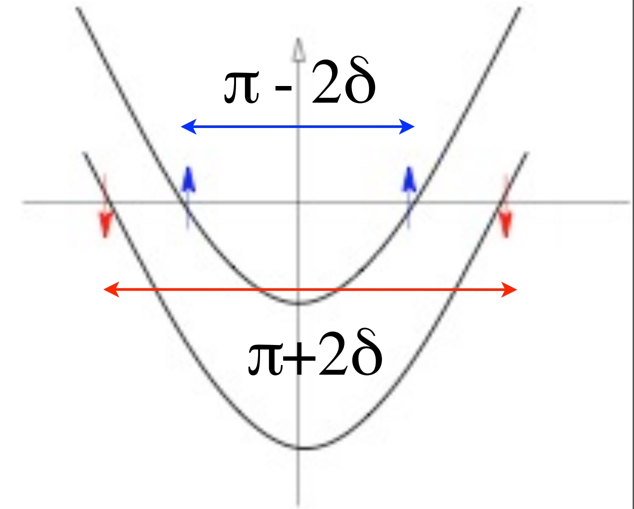
ΔS=0



1/q

- Must be careful: **often** spin-charge separation must be enforced by hand

S=1/2 AFM Chain in a Field



$$\mathcal{H} = J\sum_x \vec{S}(x)\cdot\vec{S}(x+1) - h\sum_x S^z(x)$$

- Field-split Fermi momenta:

  ✓ Uniform magnetization

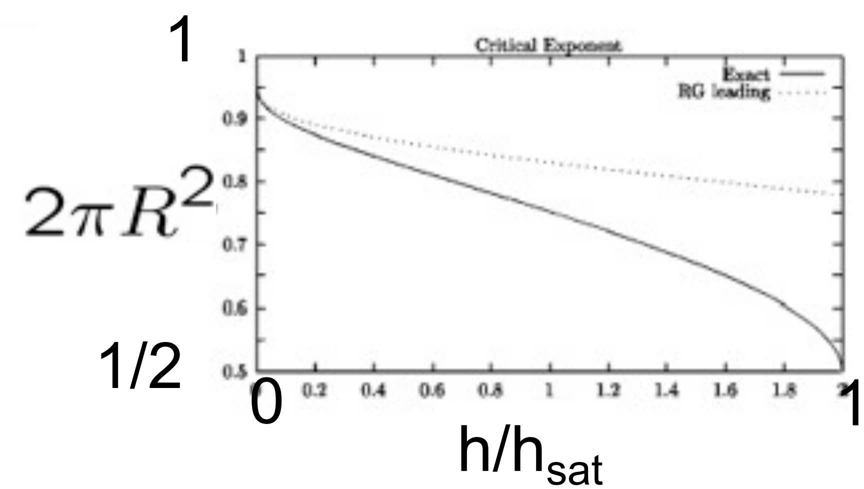
  ✓ Half-filled condition

$$k_{F\uparrow} = \frac{\pi}{2} - \delta\ ,\ k_{F\downarrow} = \frac{\pi}{2} + \delta$$

$$M = \frac{k_{F\uparrow} - k_{F\downarrow}}{2\pi} \rightarrow \delta = \pi M$$

$$n = \frac{1}{2} = \frac{k_{F\uparrow} + k_{F\downarrow}}{2\pi} \rightarrow k_{F\uparrow} + k_{F\downarrow} = \pi$$

Affleck and Oshikawa, 1999



- S^z component ($\Delta S=0$) peaked at $\pi \pm 2\delta$ scaling dimension **increases** $1/4\pi R^2$

$$S^z_{\pi\pm 2\delta}$$

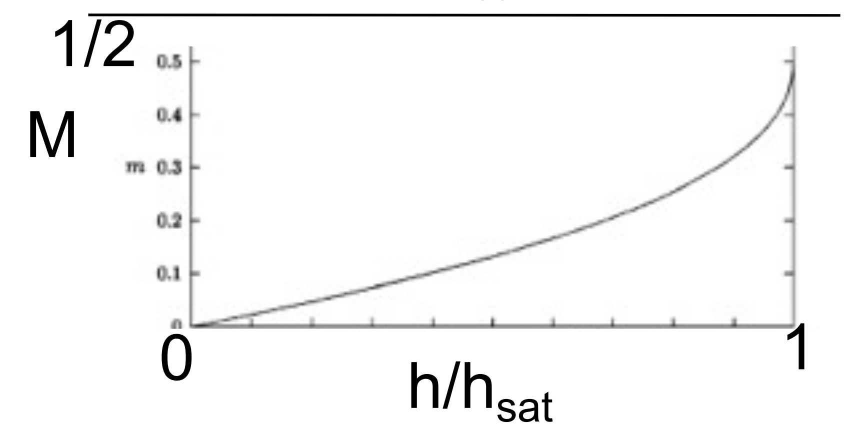
- $S^{x,y}$ components ($\Delta S=1$) remain at π scaling dimension **decreases** πR^2

$$S^{\pm}_{\pi}$$

- Derived for free electrons but correct always - *Luttinger Theorem*

1

$h_{sat}=2J$



$2k_F$ spin density fluctuations

- XY AF correlations grow with h *and remain commensurate*
- Ising “SDW” correlations decrease with h and shift from π

Ideal J-J' model in magnetic field

OS, Balents 2007

- Two important couplings for h>0
- Quantum phase transition between SDW and Cone states

Magnetic field relieves frustration!

$$\mathcal{H}_{\text{eff}} \sim \sum_{y\in 2\mathcal{Z}} \left[J' \sin(\delta) S^z_{\pi-2\delta}(y) S^z_{\pi+2\delta}(y+1) + J'\left(S^+_\pi(y)\partial_x S^-_\pi(y+1) + \text{h.c.}\right)\right]$$

dim $1/2\pi R^2$: 1 -> 2

dim $1+2\pi R^2$: 2 -> 3/2

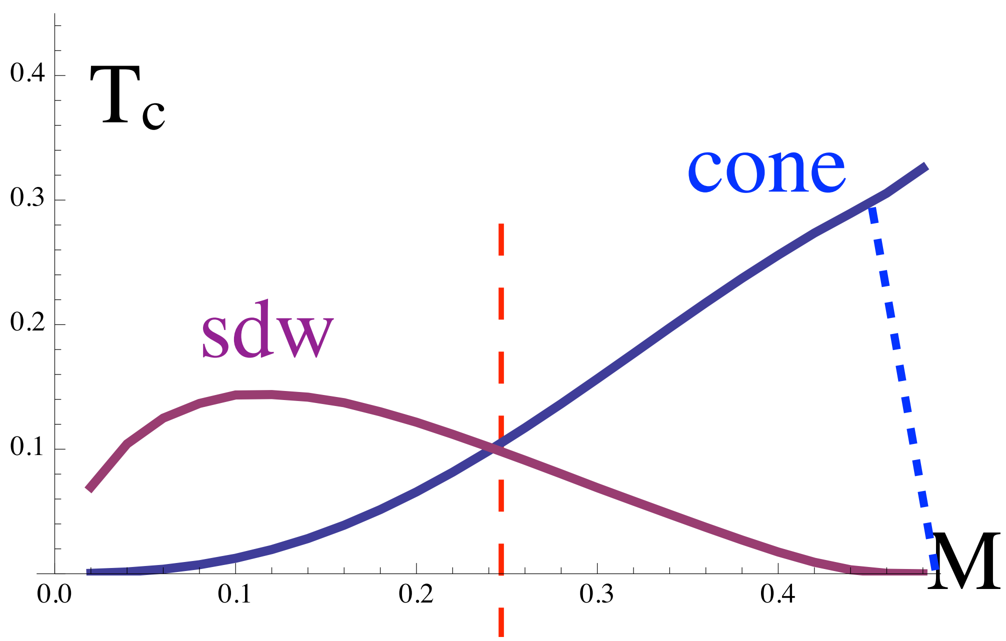
$$k_{F\downarrow} - k_{F\uparrow} = 2\delta = 2\pi M$$

"collinear" SDW

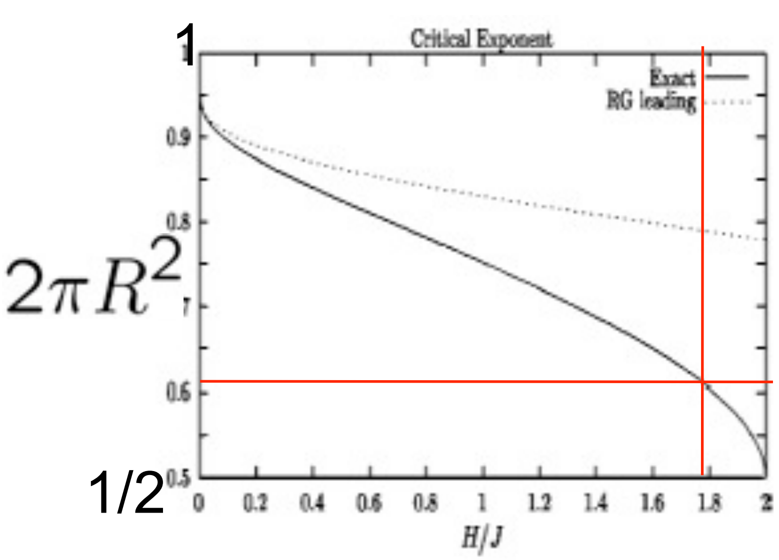
spiral "cone" state

- "Critical point": $1+2\pi R^2 = 1/2\pi R^2$ gives at M = 0.3

$$2\pi R^2 = (\sqrt{5}-1)/2 \approx 0.62$$



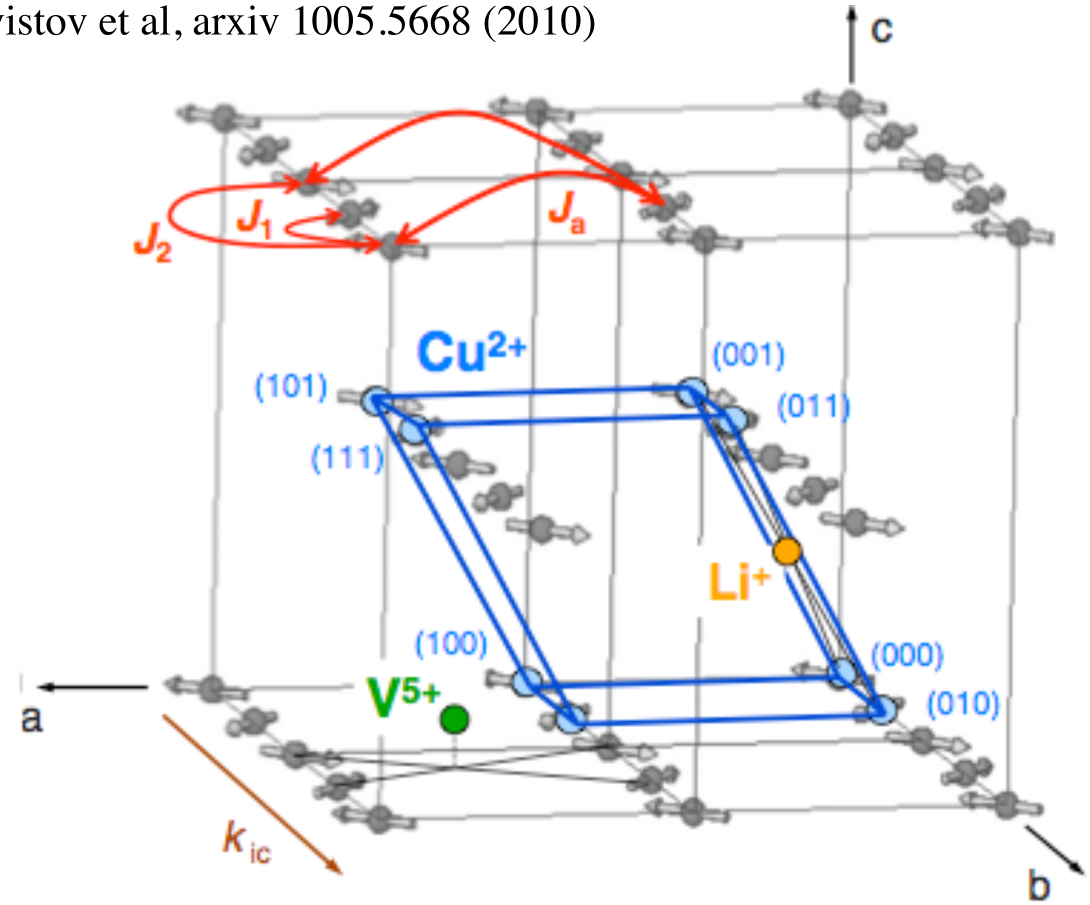
also: Kolezhuk, Vekua 2005

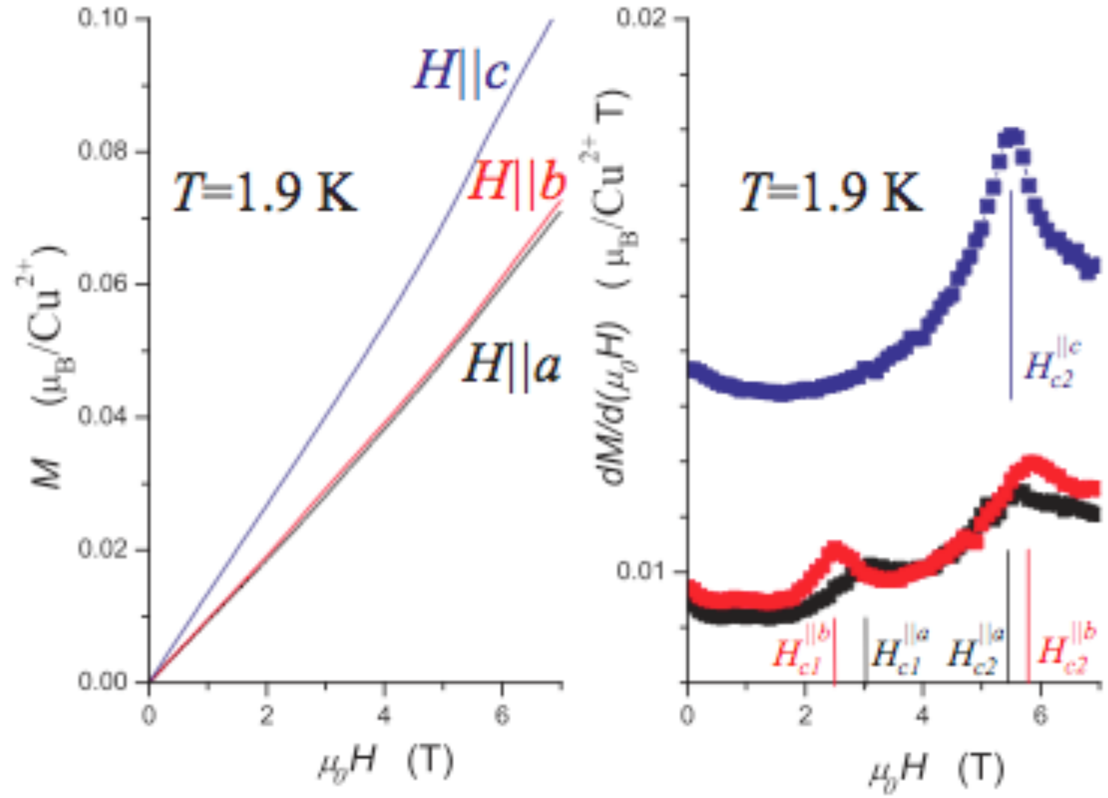


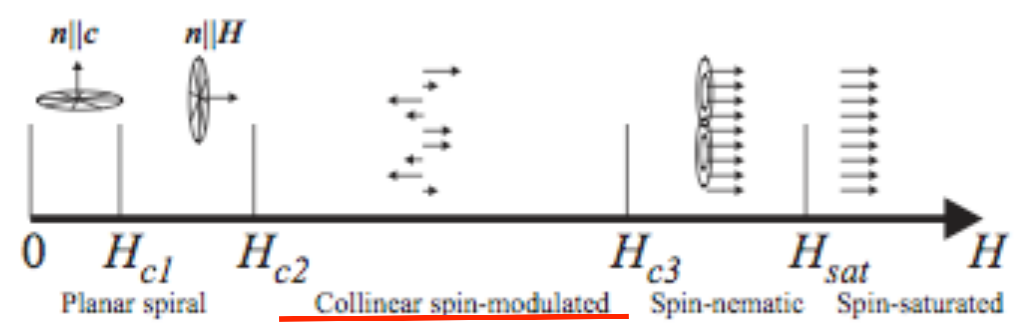
h/h_{sat}

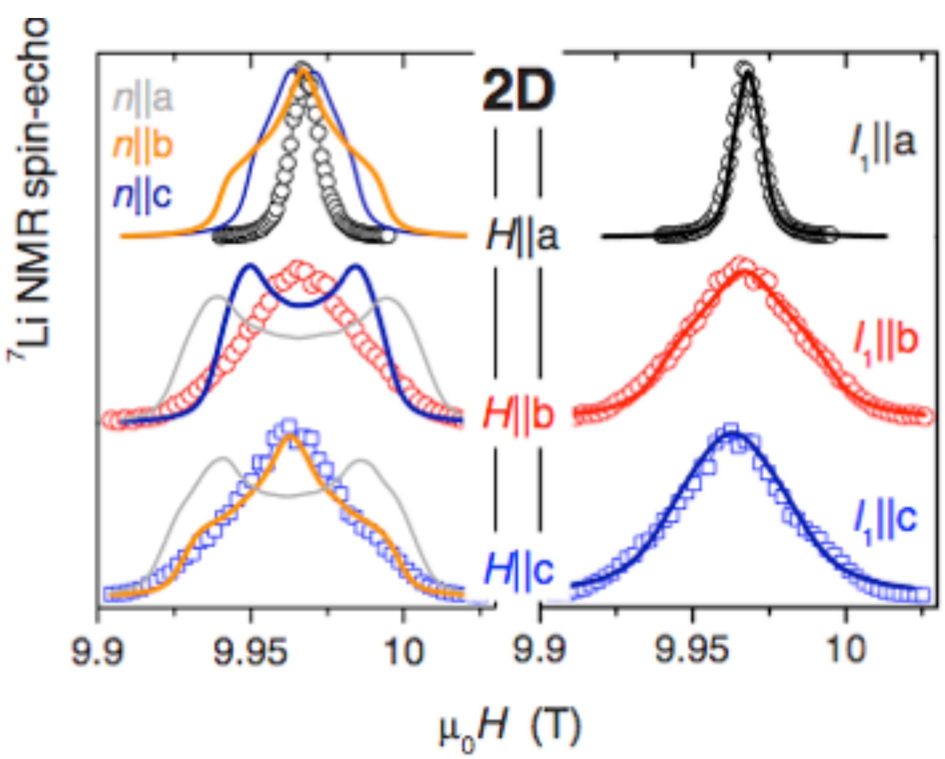
SDW in $LiCuVO_4$: J_1=-18K, J_2=49 K, J_a=-4.3K

Buttgen et al, PRB 81, 052403 (2010);
Svistov et al, arxiv 1005.5668 (2010)









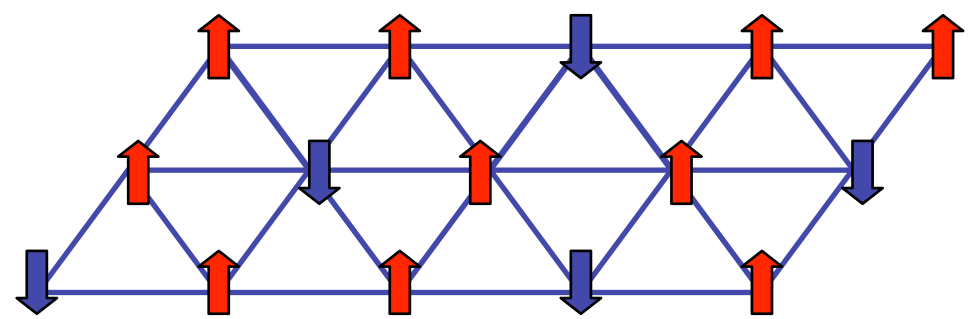
In conclusion, the magnetic structure of the high-field magnetic phase of the quasi-1D antiferromagnet $LiCuVO_4$ was studied by NMR experiments. We determined that the spin-modulated magnetic structure ($\mathbf{l}_1 \| \mathbf{H}$) with long-range magnetic order within the **ab** plane and a random phase relation between the spins of neighboring **ab** planes is realized in $LiCuVO_4$ at $H > H_{c2}$ and low temperatures $T < T_N$. The observed NMR spectra can be satisfactorily described by the following structure:

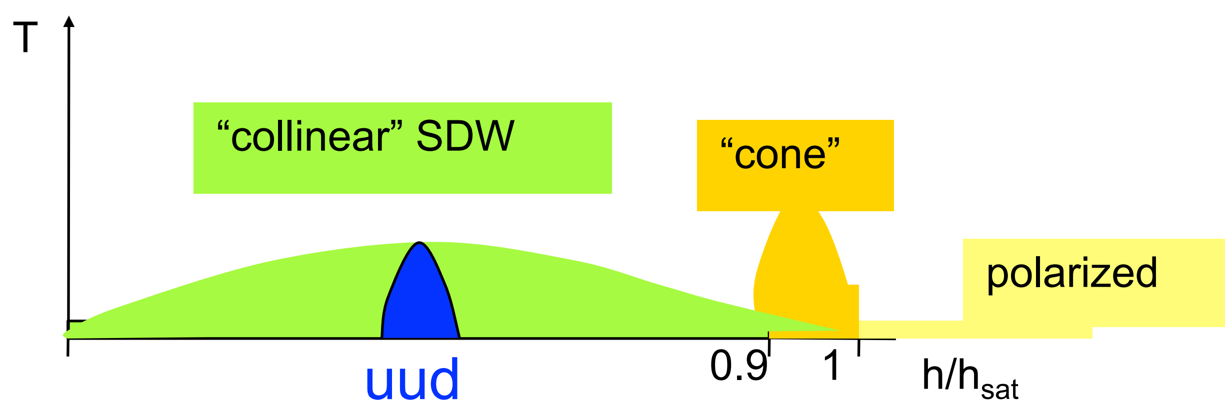
$$\mu(x,y,z) = \mu_{Cu} \cdot \mathbf{l} \cdot \cos[k_{ic} \cdot y + \phi(z)], \qquad (2)$$

where $\mathbf{l}$ is the unit vector parallel to the applied magnetic field $\mathbf{H}$ and the phase $\phi(z)$ between adjacent spins in **c** di-

J-J' model: magnetization plateaux via commensurate locking of **SDW**

- "Collinear" SDW state *locks* to the lattice at low-T
  - -"irrelevant" (1d) umklapp terms become relevant once SDW order is present (when *commensurate*): multiparticle umklapp scattering
  - -strongest locking is at M=1/3 M_{sat}
  - ✓Observed in **Cs_2CuBr_4** (Ono 2004, Tsuji 07, Fortune 09)
    - ***down***-spins at the centers of hexagons





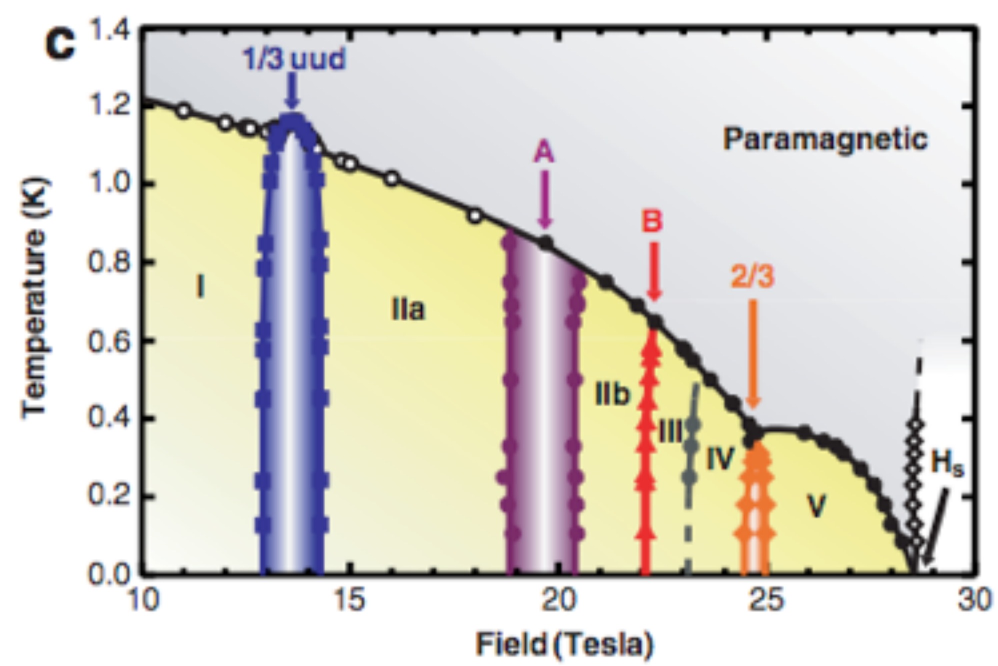
$$\left(\Psi_R^\dagger \Psi_L\right)^n \rightarrow (\pi - 2\delta)n = 2\pi m \rightarrow \boxed{2M = 1 - 2m/n}$$

n	3	4	5	5	6
m	1	1	1	2	1
2M	1/3	1/2	3/5	1/5	2/3

naively thinking

Cs_2CuBr_4 Fortune et al 2009

1/3 2/3



Plateau more carefully

OS, Katsura, Balents PRB 2010

$$M^{(n,m)} = \frac{1}{2}\left(1 - \frac{2m}{n}\right)$$

- Umklapp *must* respect triangular lattice symmetries
  - translation along chain direction $\phi_y(x) \to \phi_y(x+1) - R(\pi - 2\delta)$
  - translation along diagonal $\phi_y(x) \to \phi_{y+1}(x+1/2) - R(\pi - 2\delta)/2$
  - spatial inversion $\phi_y(x) \to \pi R - \phi_y(-x)$

$$H^{(n)}_{umk} = \sum_y \int dx\, t_n \cos[\frac{n}{R}\phi_y]$$ **and** $$n = m \ (mod\ 2)$$ ***same parity condition***

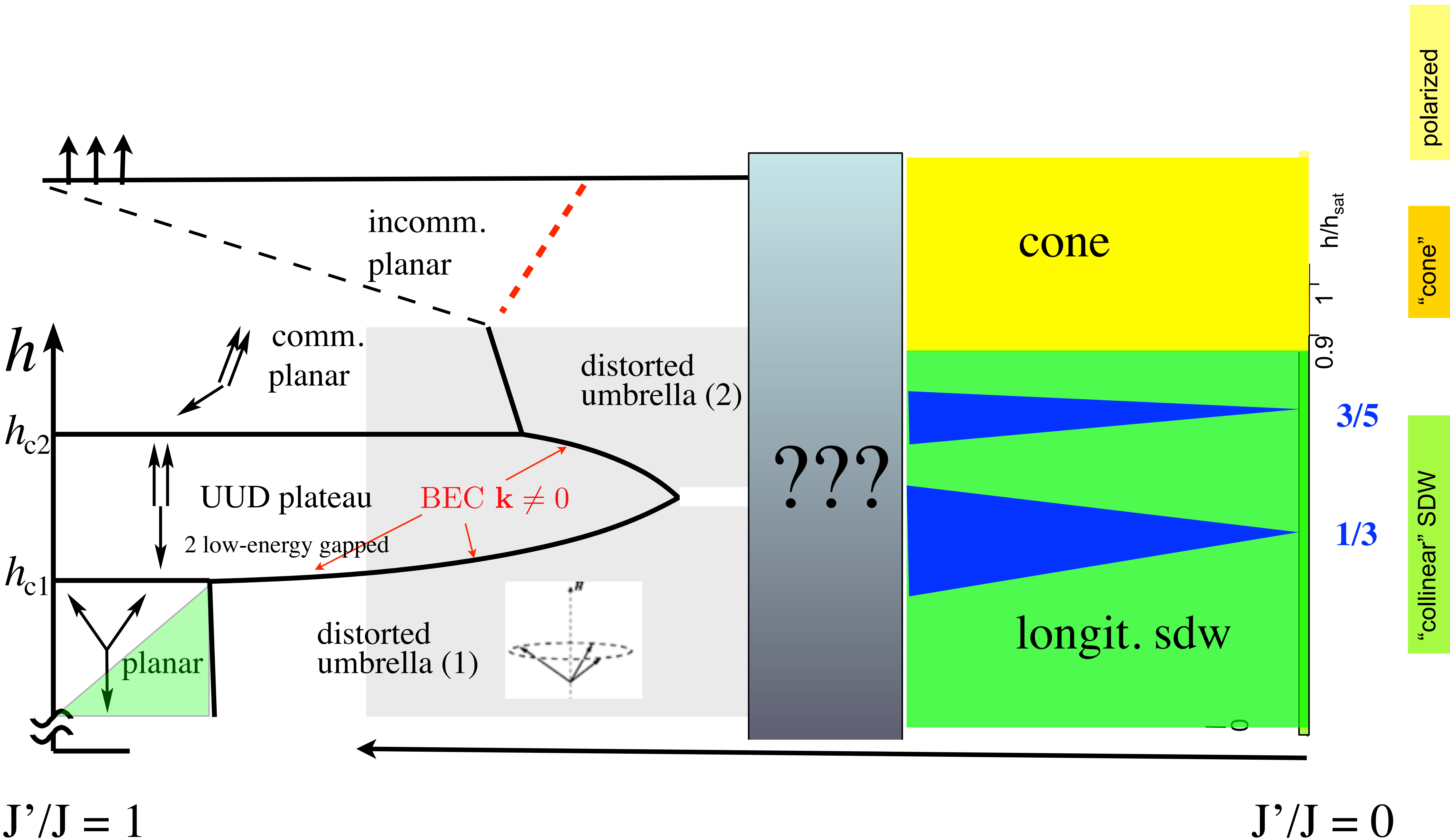
- **n**-th plateau width (in field) $width \sim \left(J'/J\right)^{n^2/(4(4\pi R^2-1))}$

n	**3**	**8**	**5**	**10**	**12**
m	**1**	**2**	**1**	**4**	**2**
2M	**1/3**	1/2	**3/5**	1/5	2/3

large **n** leads to exponential suppression

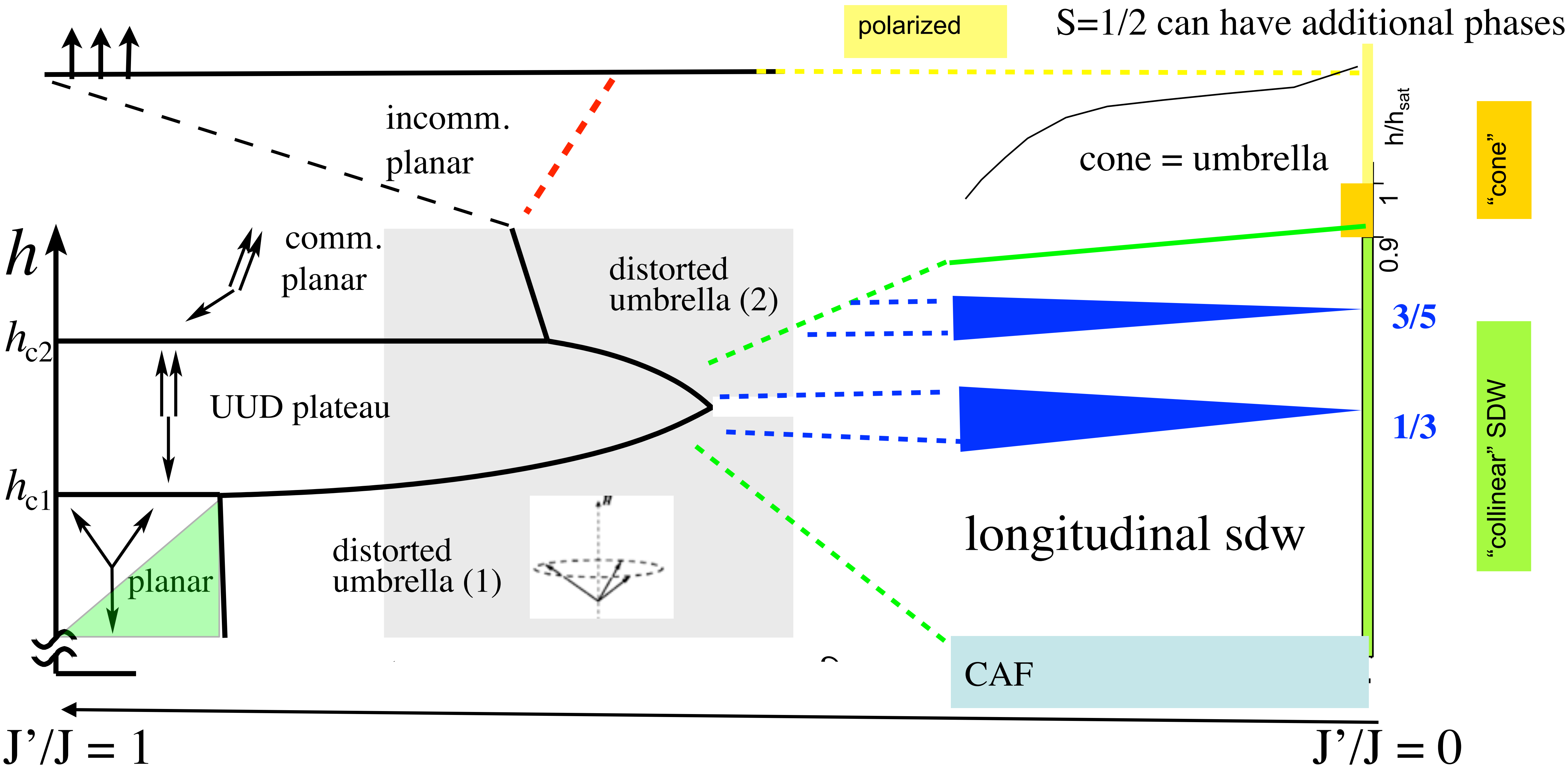
- 1/3-plateau is most prominent, 3/5 is possible (if falls within the SDW region). Exponentially weak 1/2- and 2/3- plateaux, if any !

How to interpolate J’ << J limit to J’ = J point?

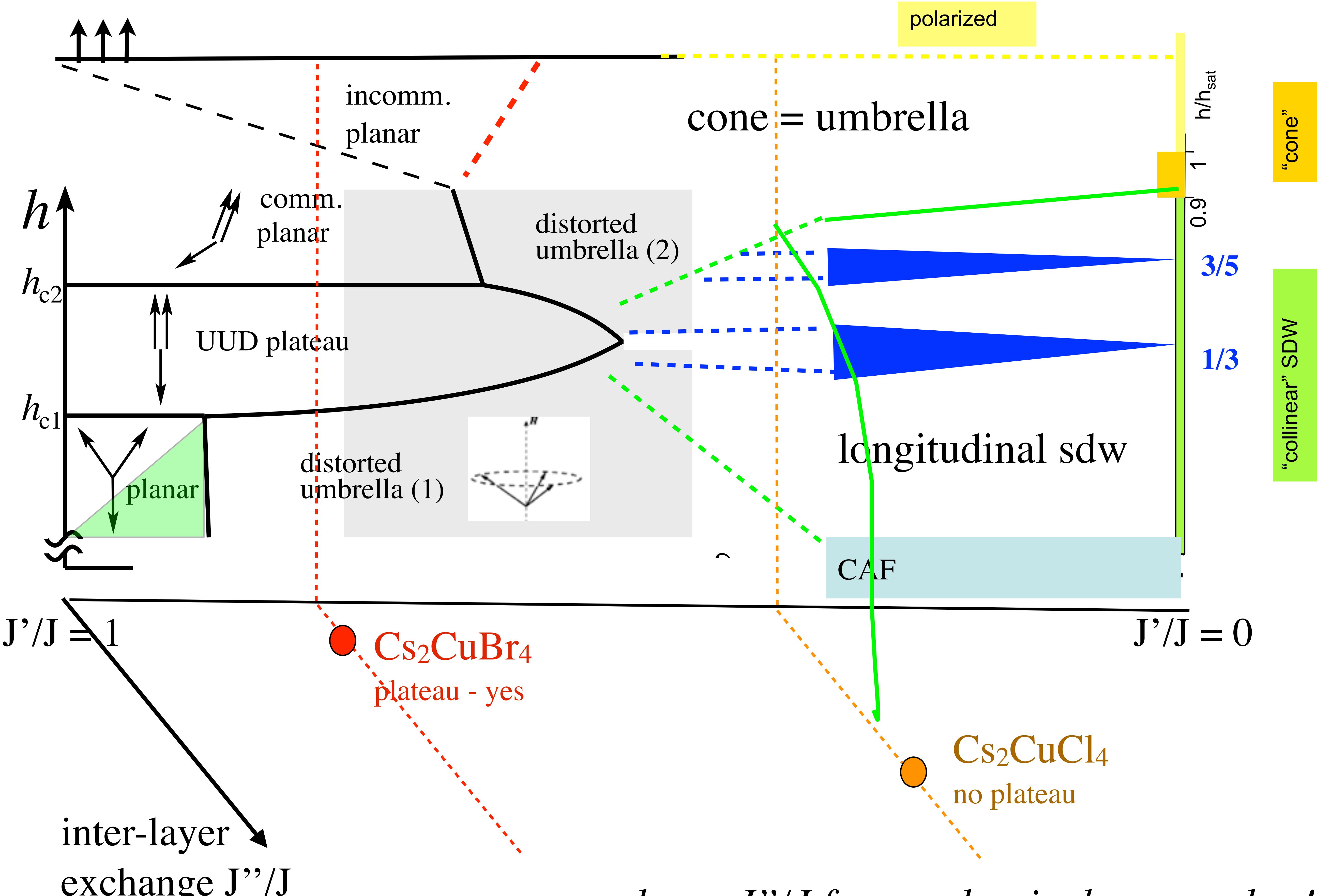


Global phase diagram

Hypothesis: 1/3 plateau extends for all 0 < J'/J < 1;
other magnetization plateaux terminate above some critical J'/J ratio.



Experimental relevance



large J''/J favors classical cone order !

Conclusions

- Large degeneracy of classical triangular lattice antiferromagnet
- Planar and UUD phases selected by quantum/ thermal fluctuations
- Magnetization plateau persists all way to weakly coupled chains
- Other interesting instabilities of collinear SDW?