Spinon spin resonance

~~Electron~~ spin resonance of spinon gas

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In collaboration with:

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A. Shapiro, Shubnikov Institute for Crystallography, Moscow, Russia

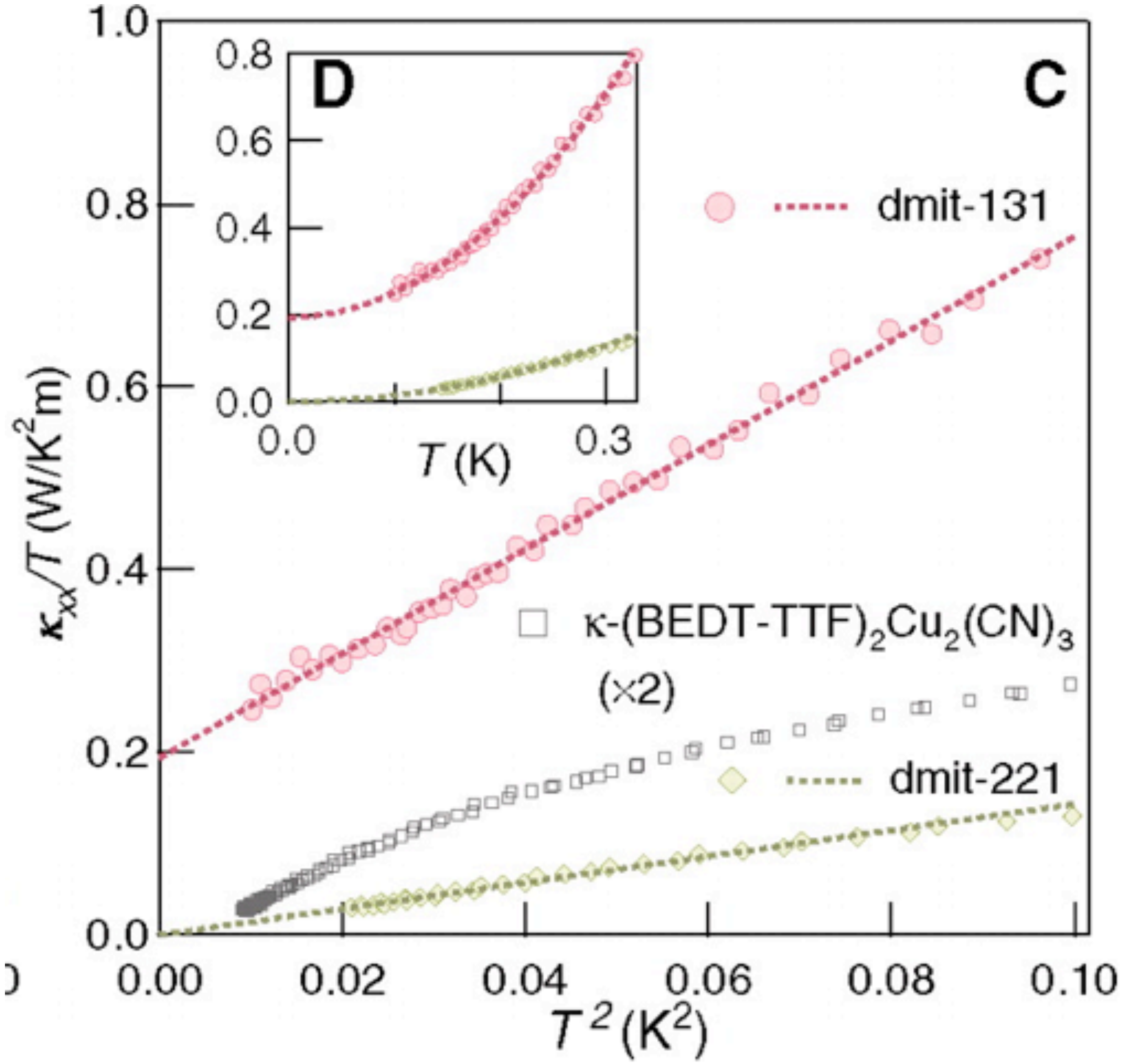
Rachel Glenn and **Mikhail Raikh** (U Utah)

Exotic phases of frustrated magnets, KITP, October 8-12, 2012

Outline

- *Main ingredients*
  - spin liquid with spinon Fermi surface
  - DM interaction
- *Case study* Cs_2CuCl_4: Spinon continuum and ESR
- *Higher dimensional extension*: ESR in the presence of uniform DM interaction
  - from Hubbard to spin liquid with spinon Fermi surface
- Conclusions

Spin liquid with spinon Fermi surface



M. Yamashita et al, Science 2010

theory: O. Motrunich 2005,
S.-S. Lee and P. A. Lee 2005

electrical insulator,
metal-like thermal conductor

Amusing bit of history: I. Pomeranchuk, J. Phys. USSR vol. 4 pp. 357-374 (1941)

THE THERMAL CONDUCTIVITY OF THE PARAMAGNETIC DIELECTRICS AT LOW TEMPERATURES

By I. POMERANCHUK

(Received October 25, 1940)

In the paramagnetic dielectrics the paramagnetic spectrum can take part in the heat transfer. Besides the mutual collision (as in the ordinary dielectrics) the free path of the phonons essentially depends on this spectrum. At low temperatures the main part in the conduction of heat is played in some cases by the phonons, in the others by the paramagnetic spectrum. The run of the curve of termal conductivity as function of temperature has a complicated character with a number of maxima and minima, and is quite different as compared with the corresponding situation in ordinary dielectrics. An experimental check of the theory could give opportunity to find out the character of the paramagnetic exchange spectrum.

Spin liquid with spinon Fermi surface

Regarding the nature of excitations:

...the magnetic excitations levels correspond to the deviations from the normal distribution of the magnetic moments which are propagating through the whole crystal and are not localized in a definite place of the lattice. Such magnetic excitations will be called in the following "**magnons**"
(**this name was suggested by L. Landau**).

Regarding statistics of spin excitations:

“The experimental facts available suggest that the magnons are submitted to the Fermi statistics; namely, when $T << T_{CW}$ the susceptibility tends to a constant limit, which is of the order of const/T_{CW} ([5]) [for $T > T_{CW}$, χ=const/($T + T_{CW}$)]. Evidently we have here to deal with the Pauli paramagnetism which can be directly obtained from the Fermi distribution. Therefore, we shall assume the Fermi statistics for the magnons.”

Ref. (5) A. Perrier and Kamerlingh Onnes, Leiden Comm. No.139 (1914)

Dzyaloshinskii-Moriya (DM) interaction

D

J. Phys. Chem. Solids Pergamon Press 1958. Vol. 4. pp. 241–255.

A THERMODYNAMIC THEORY OF "WEAK" FERROMAGNETISM OF ANTIFERROMAGNETICS

I. DZYALOSHINSKY
Institute for Physical Problems, Academy of Sciences of the U.S.S.R., Moscow

(*Received* 19 *February* 1957)

M

PHYSICAL REVIEW VOLUME 120, NUMBER 1 OCTOBER 1, 1960

Anisotropic Superexchange Interaction and Weak Ferromagnetism

TÔRU MORIYA*
Bell Telephone Laboratories, Murray Hill, New Jersey

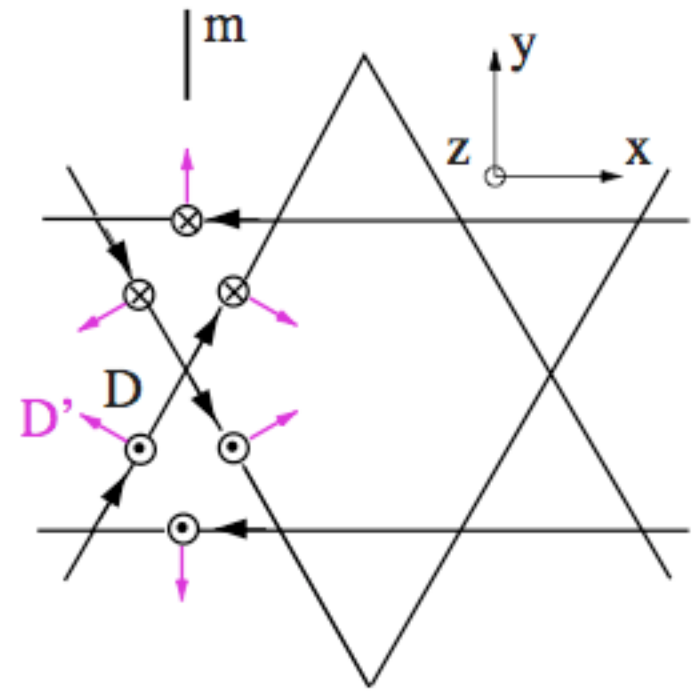
(Received May 25, 1960)

$$\mathbf{D}_{ij} \cdot \mathbf{S}_i \times \mathbf{S}_j$$

- Reduces symmetry to U(1) [rotations about D axis]
- Easy plane anisotropy (perp. to D)
- Promotes magnetic order
- Generically stabilizes incommensurate non-collinear (spiral) states

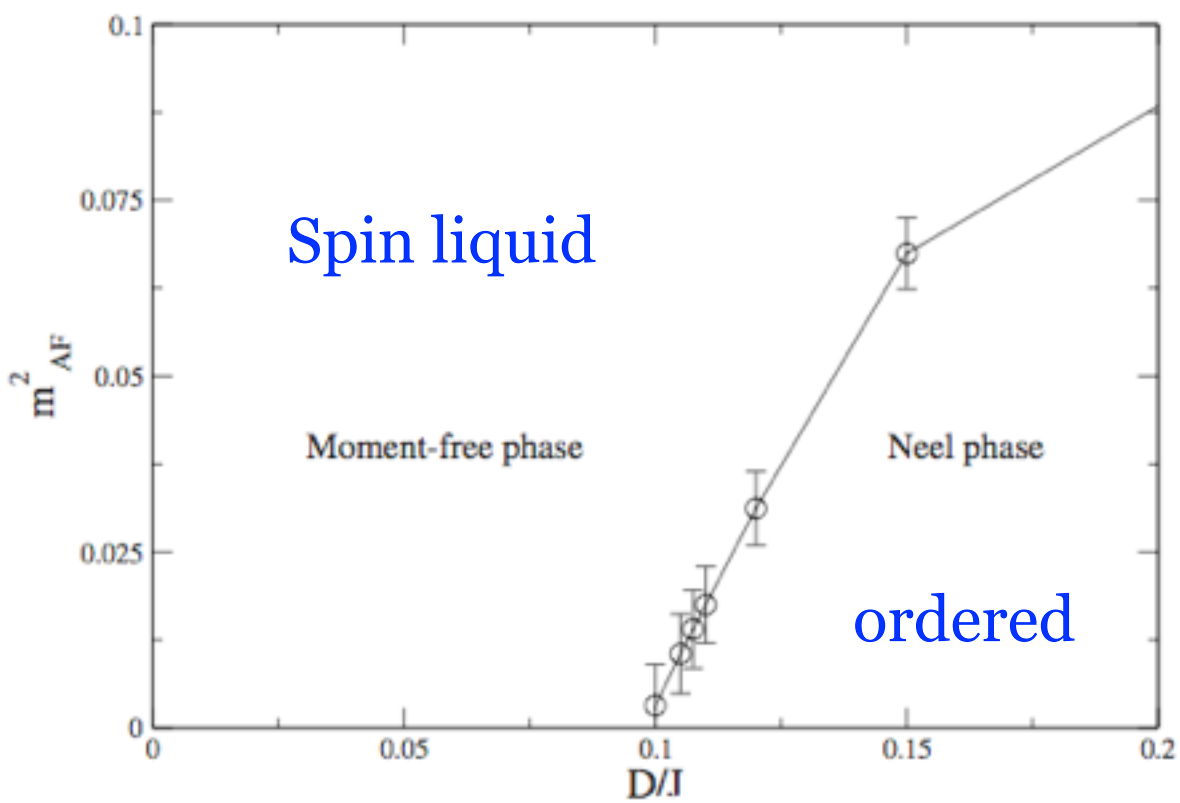
Example: DM in kagome

main DM: orthogonal to kagome plane; staggered between up and down triangles



$$H = \sum_{nn} [J\mathbf{S}_i \cdot \mathbf{S}_j + \mathbf{D}_{ij} \cdot (\mathbf{S}_i \times \mathbf{S}_j)]$$

Elhajal, Canals, and Lacroix, PRB 2002
Rigol, Singh 2007



Cepas, Fong, Leung, Lhuillier, PRB 2008

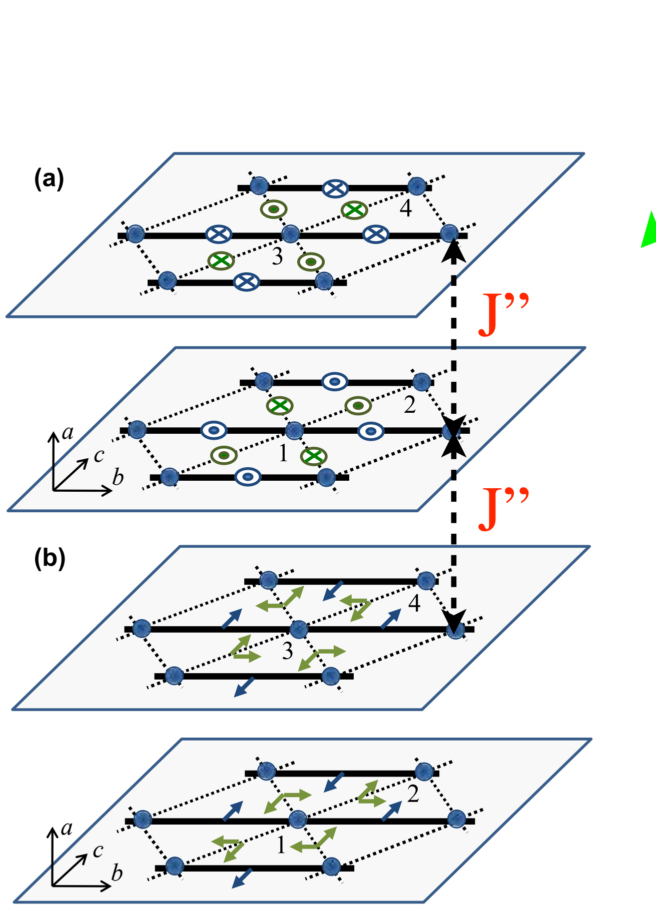
Main idea:

- turn annoying material imperfection (DM) into a probe of exotic spin state and its excitations
- probe small-**q** excitations by ESR

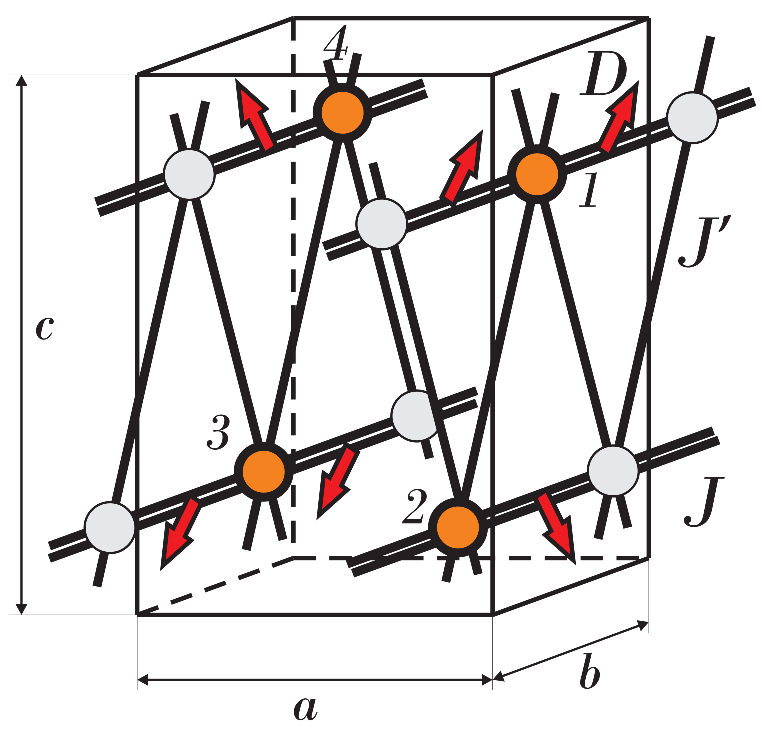
Cs_2CuCl_4: consequences of Dzyaloshinskii-Moriya interaction

$$\vec{D}_{ij} \cdot \vec{S}_i \times \vec{S}_j$$

- Is known from inelastic neutron scattering data (Coldea et al. 2001-03)
- 3D ordered state - determined by weak residual interactions - interplane and ***Dzyaloshinskii-Moriya*** (DM) OS, Katsura, Balents 2010



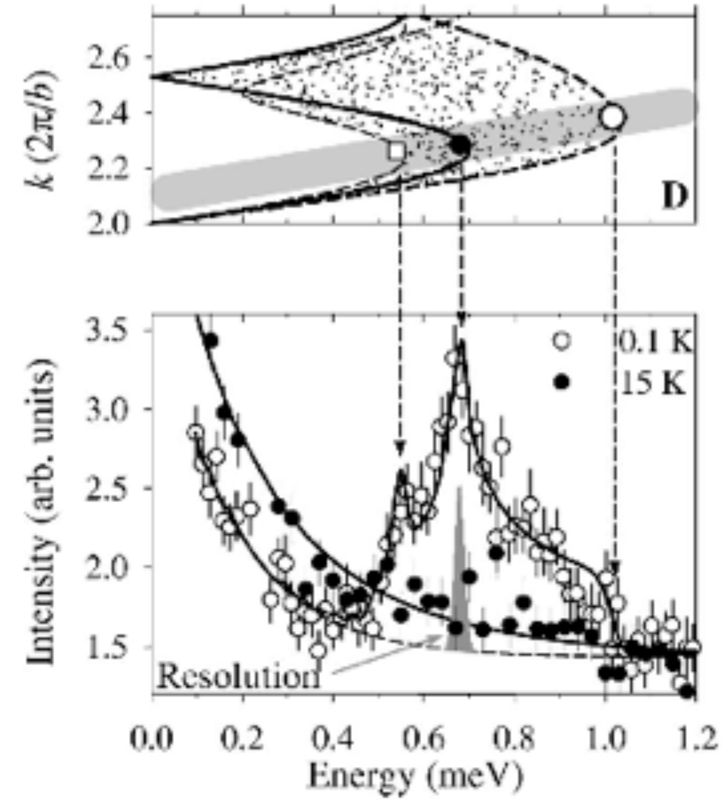
5 different DM terms allowed



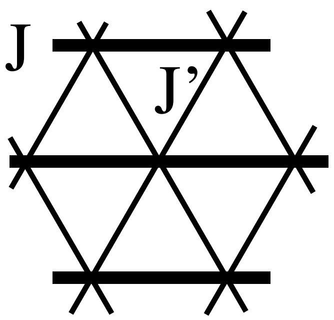
VOLUME 86, NUMBER 7 PHYSICAL REVIEW LETTERS 12 FEBRUARY 2001

Experimental Realization of a 2D Fractional Quantum S

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



PHYSICAL REVIEW B 68, 134424 (2003)



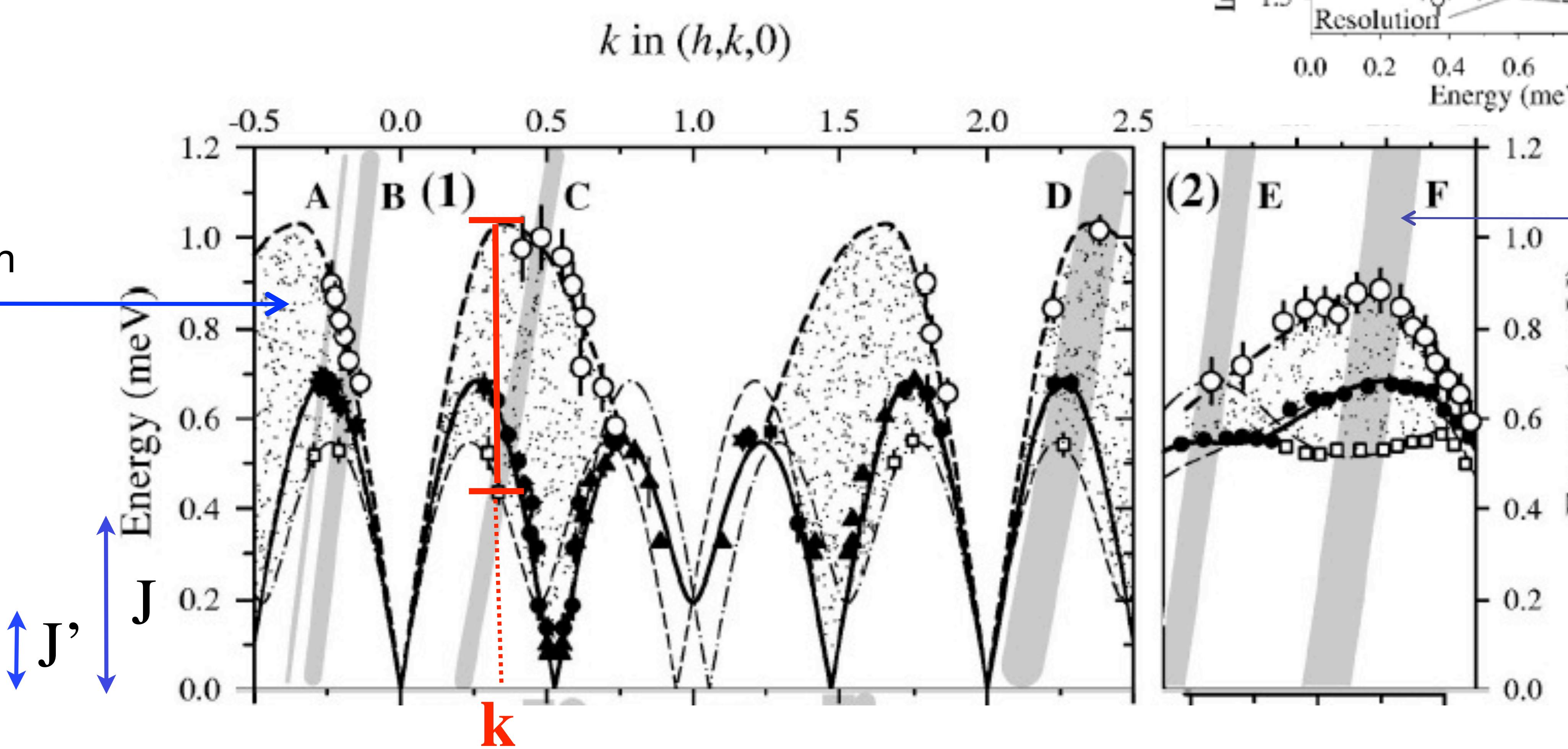
Extended scattering continua characteristic of spin fractionalizatio frustrated quantum magnet Cs_2CuCl_4 observed by neut

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

Along the chain

Cs_2CuCl_4

$J'/J=0.34$

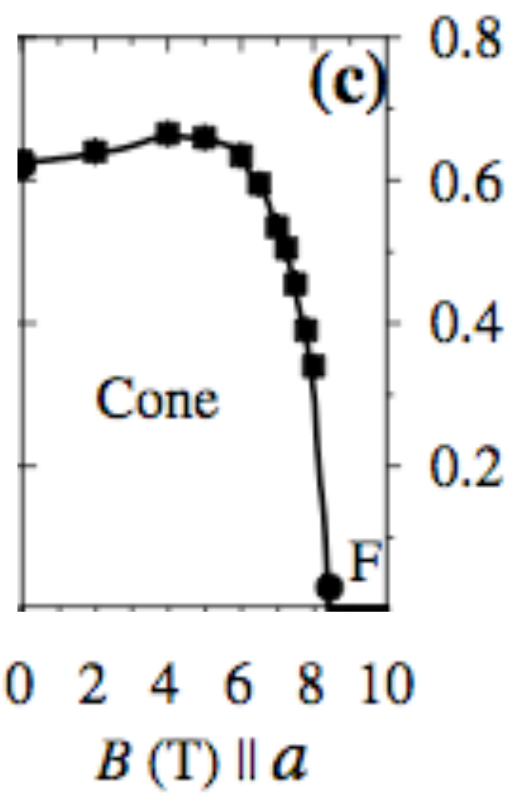


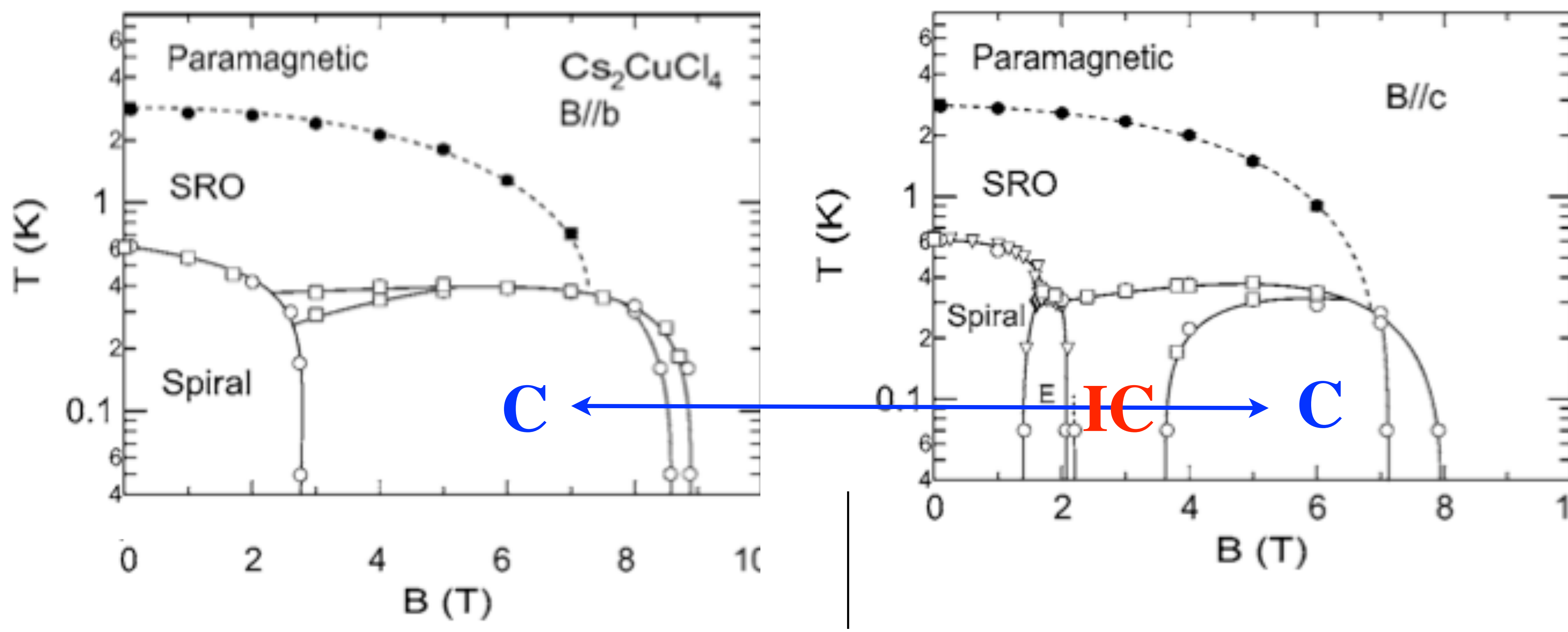
transverse to chain

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

Highly anisotropic phase diagram of Cs_2CuCl_4 in magnetic field is explained by DM interactions

Tokiwa et al, 2006
Veillette, Chalker, Coldea 2005
Starykh, Katsura, Balents 2010

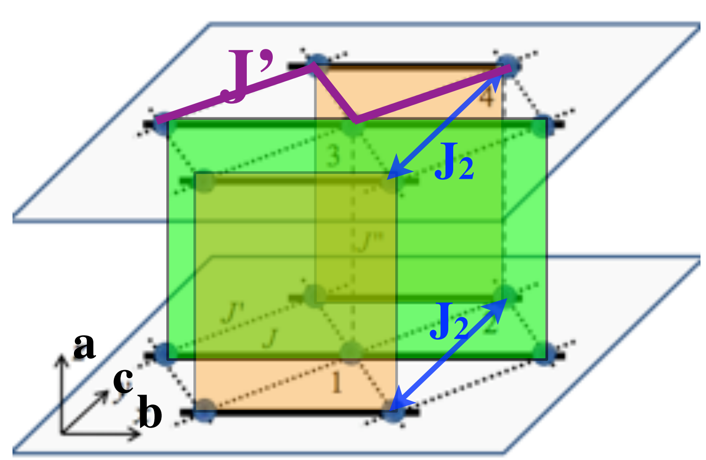




DM controlled cone state

$$(-1)^z D'_a \hat{a}$$

B || b
DM free situation



B || c
DM driven
C-IC transition

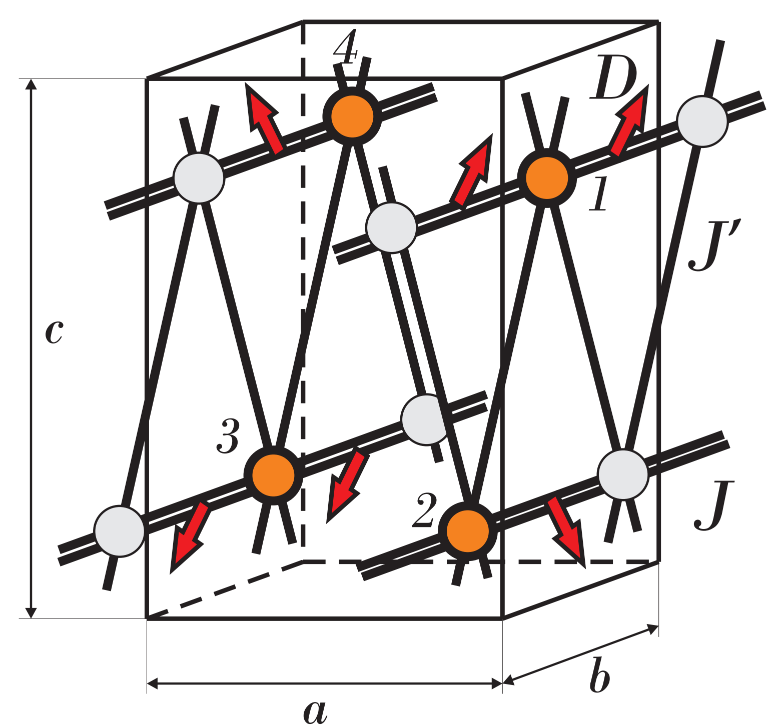
$$(-1)^y D_c \hat{c}$$

Cs_2CuCl_4: uniform Dzyaloshinskii-Moriya interaction

$$\vec{D}_{ij} \cdot \vec{S}_i \times \vec{S}_j$$

Focus on the in-chain DM: for a given chain (y,z) vector D is constant

$$D_{y,z} = (-1)^y D_c \hat{c} + (-1)^z D_a \hat{a}$$



ESR - electron spin resonance

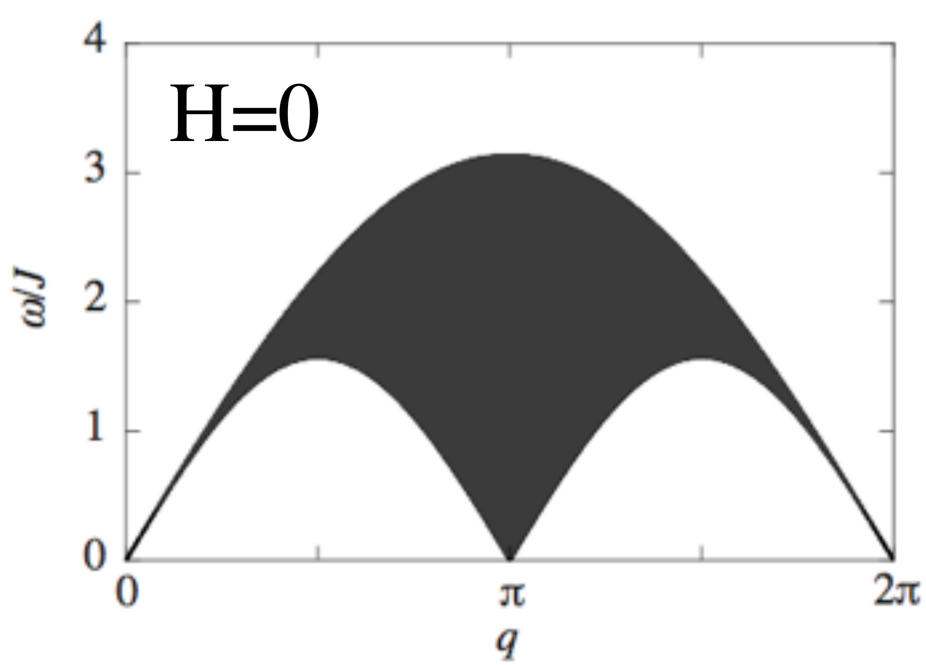
- Simple (in principle) and sensitive probe of magnetic *anisotropies* (and, also, **q=0** probe: $S = \Sigma_r S_r$)

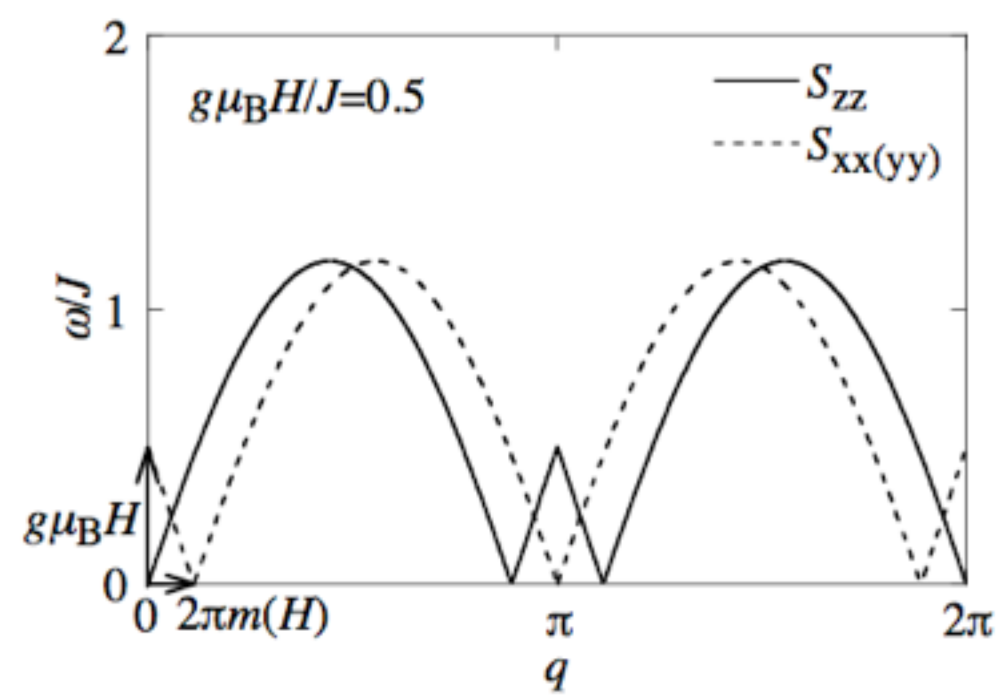
$$I(h,\omega) = \frac{\omega}{4L} \int dt e^{i\omega t} \langle [S^+(t), S^-] \rangle$$

Zeeman **H** along **z**, microwave radiation **h** polarized perpendicular to it.

- For SU(2) invariant chain in paramagnetic phase

I(H,ω) ~ δ(ω-H) m(H) [Kohn's Th] Oshikawa, Affleck PRB 2002

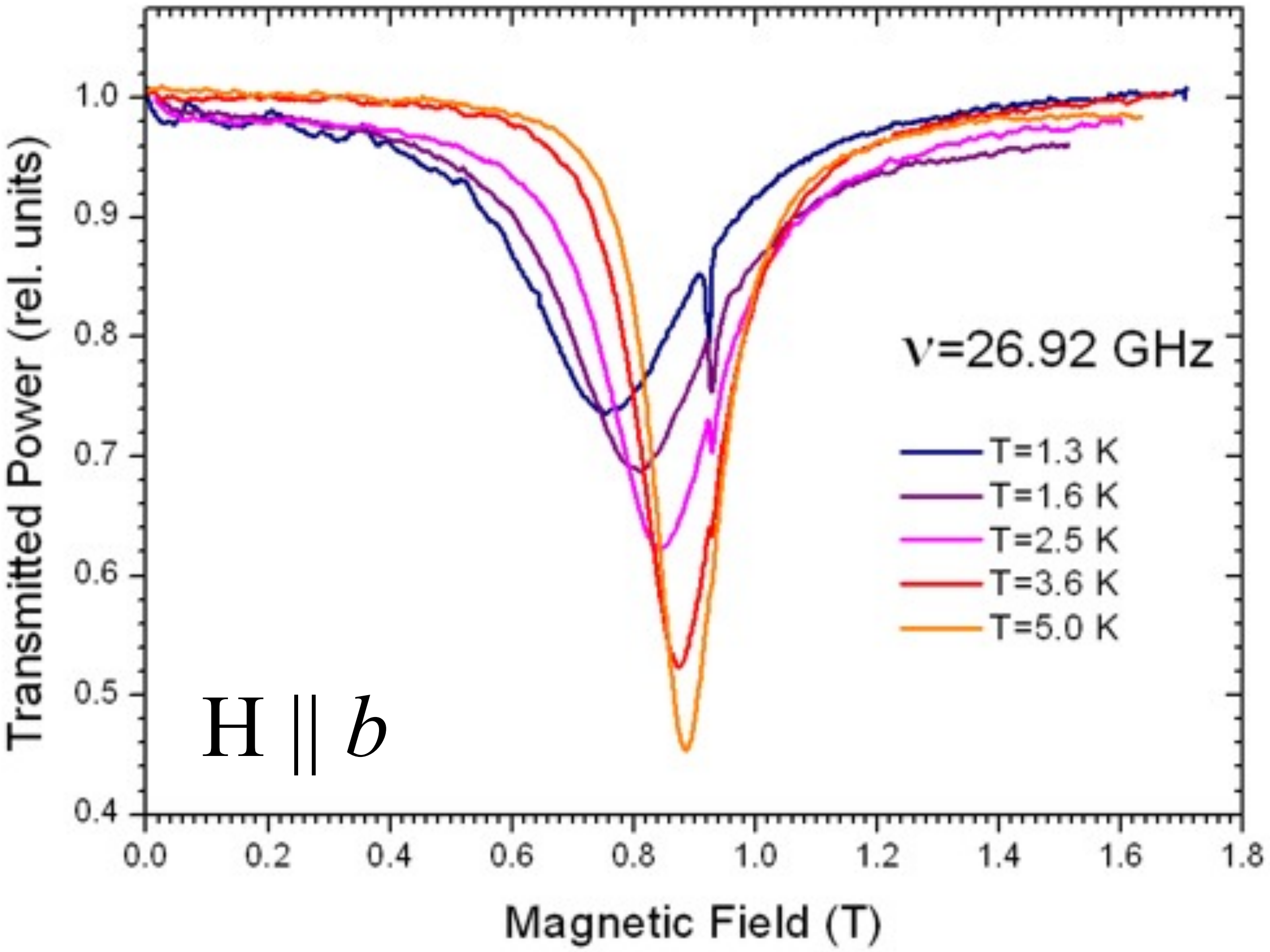


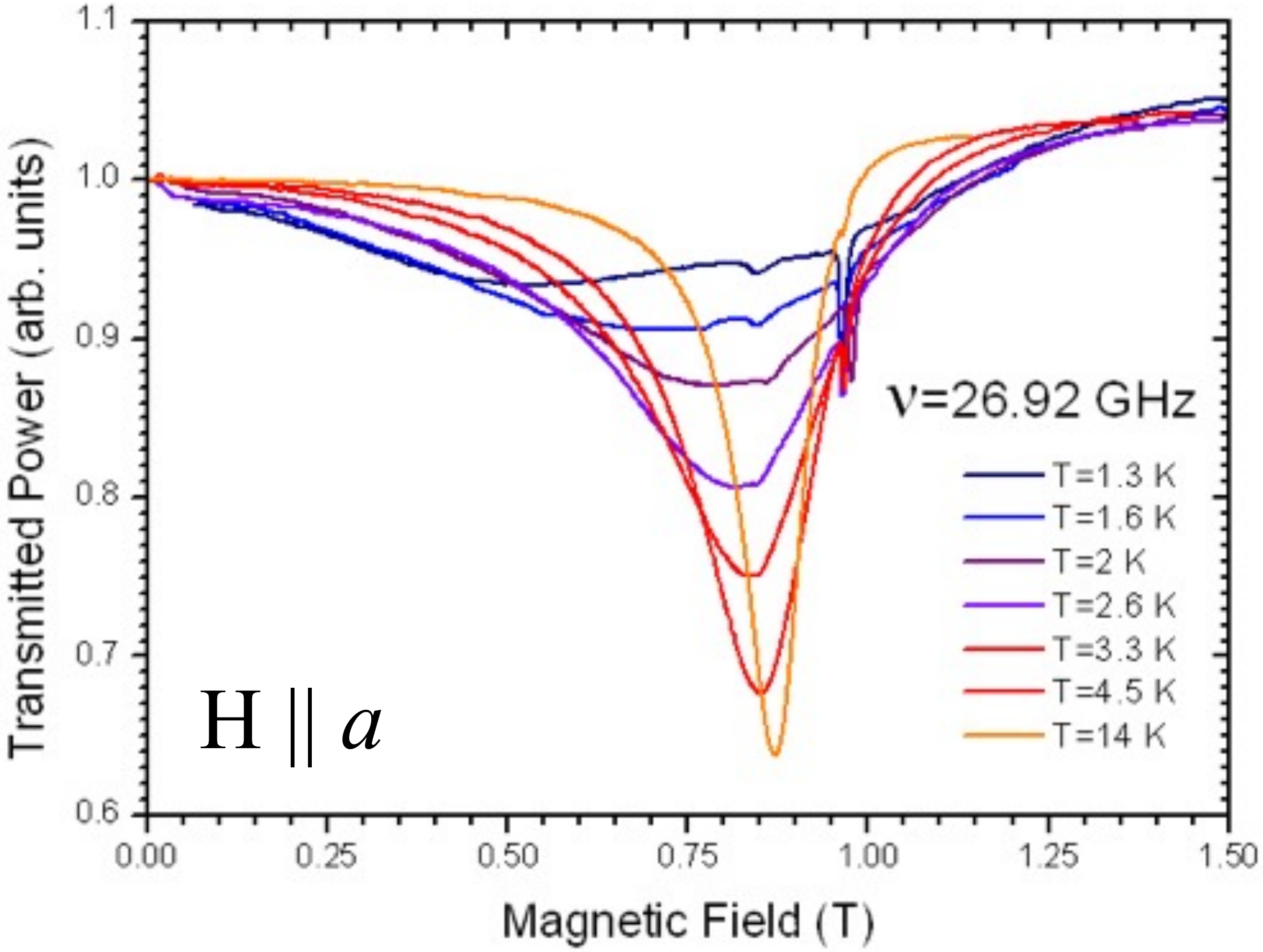


ESR data (Povarov and Smirnov, Kapitza Institute)

Shift for H || *b*

Shift and splitting for H || *a*, *c*

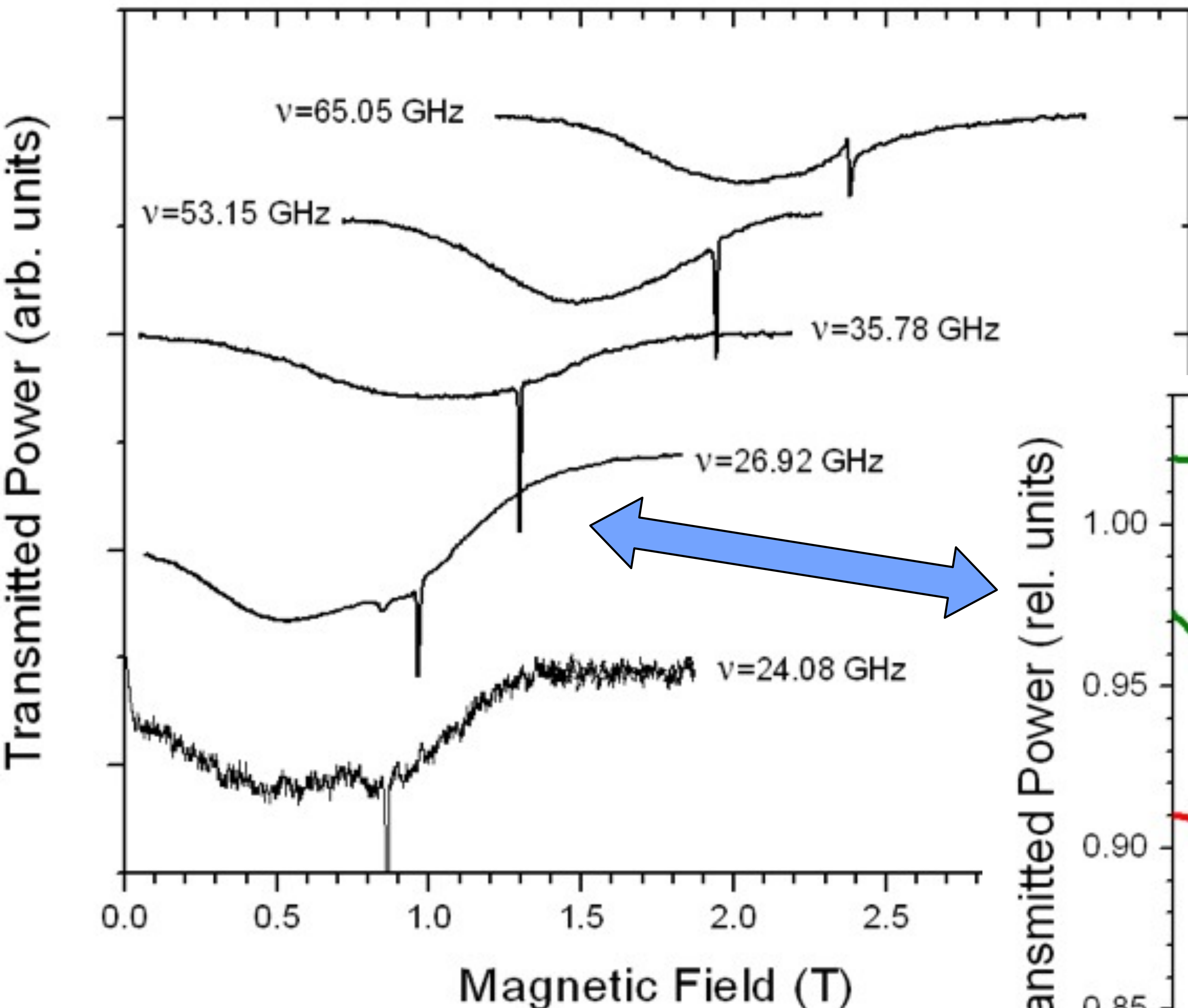




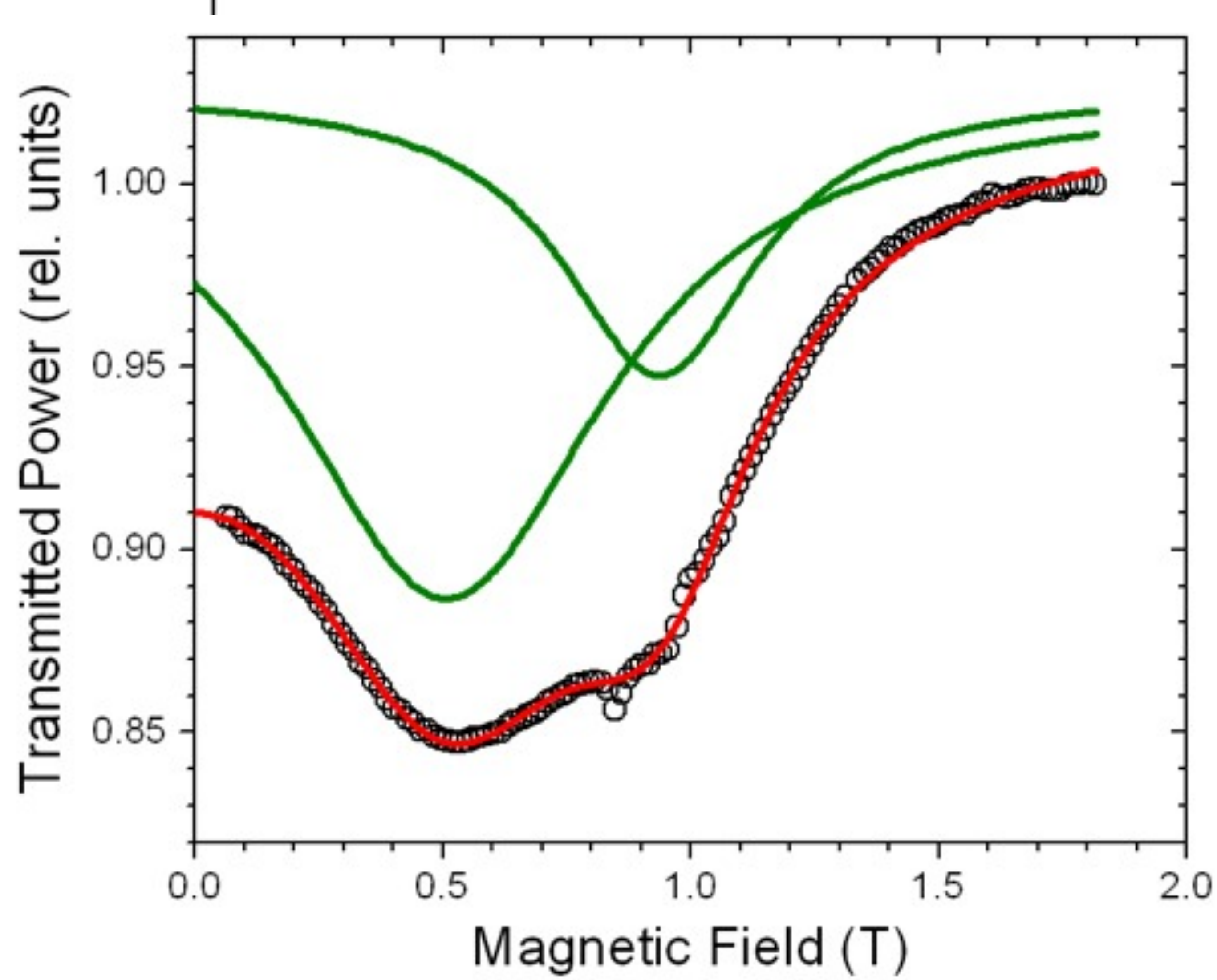
Resonance line is significantly modified with lowering the temperature; modification is strongly anisotropic with respect to field.

The lowest temperature T=1.3 K is still twice higher than ordering T_N

Line splitting



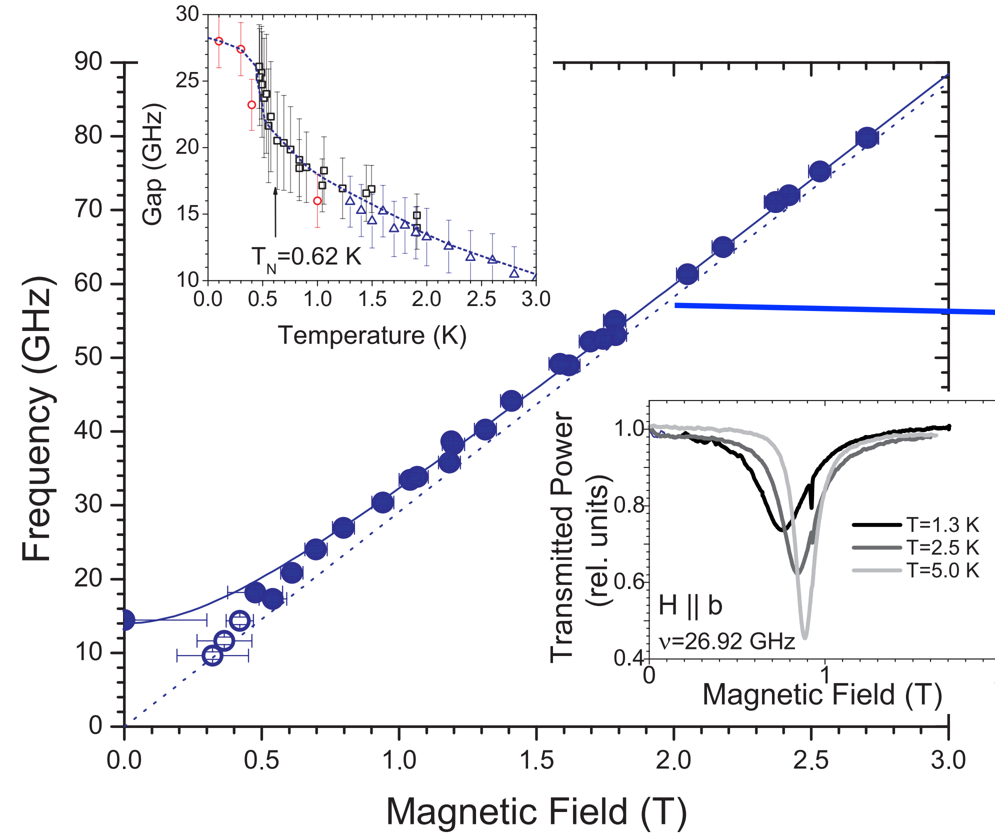
T=1.3 K
ν=26.92 GHz
H || a



Cs_2CuCl_4 ESR data: T=1.3 K

Povarov et al, 2011

- H along b-axis

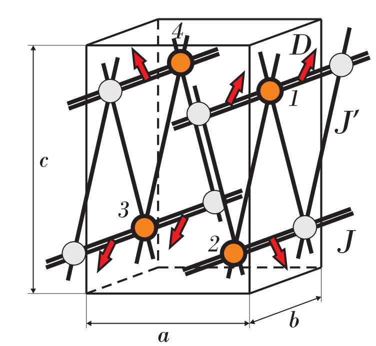


$g_b = 2.08$

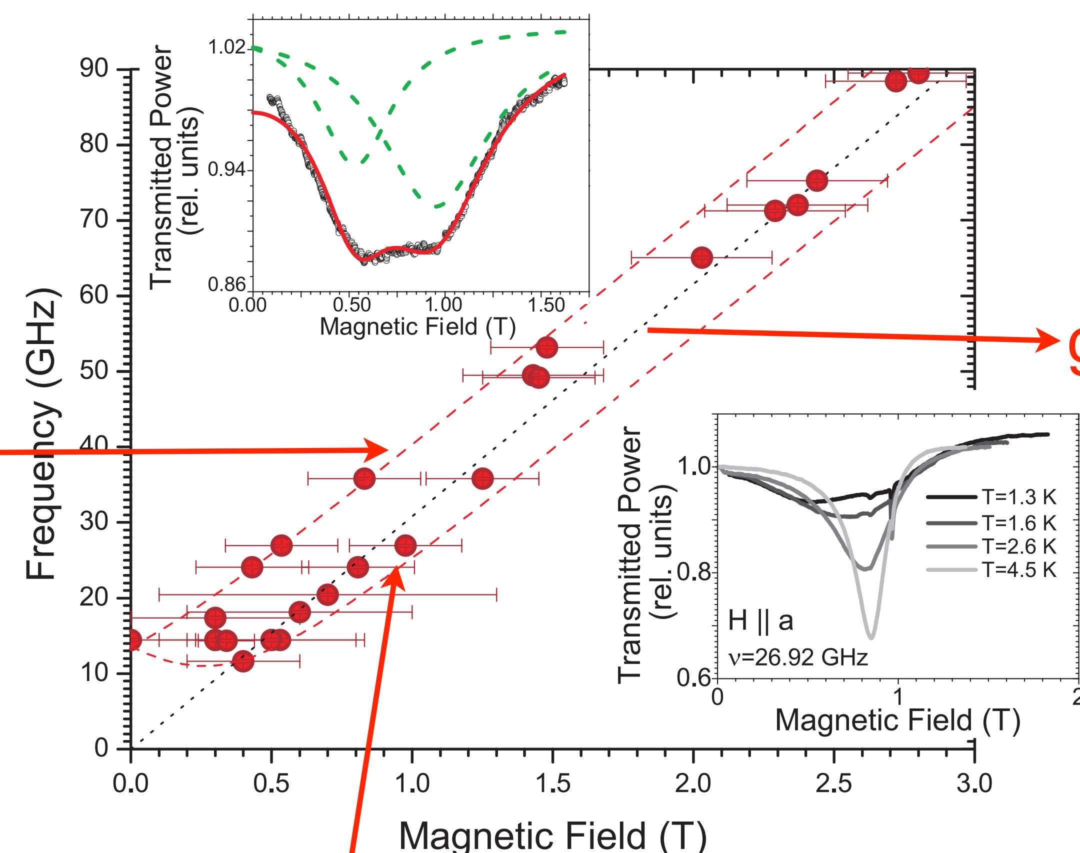
✓ **gap**-like behavior for ν > 17 GHz

$$2\pi\hbar\nu = \sqrt{(g_b\mu_B H)^2 + \Delta^2}$$

✓ loss of intensity for ν < 17 GHz



- H along a-axis: splitting of the ESR line



$g_a = 2.20$

Previous experiments

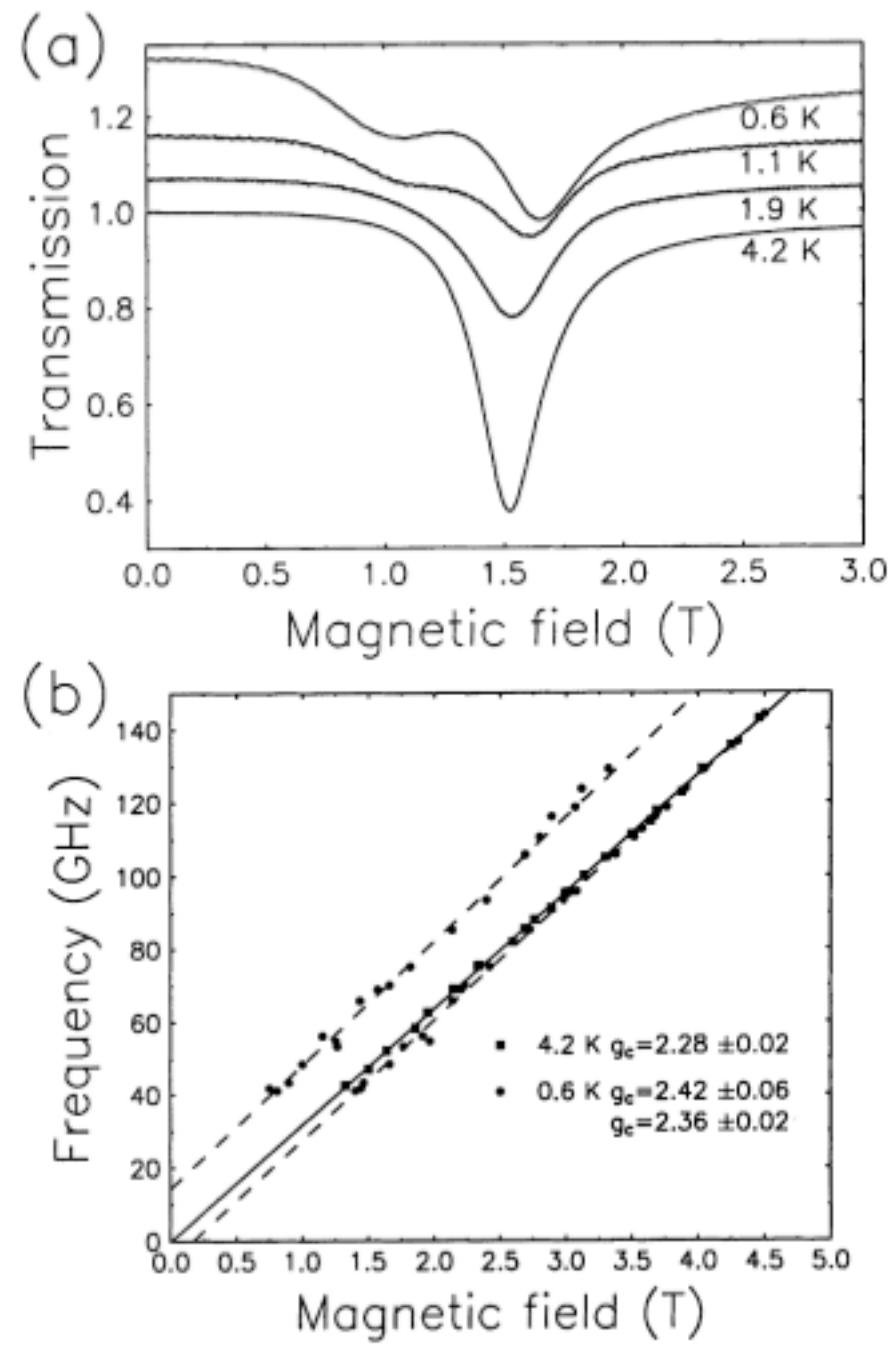
Physica B 256-258 (1998)

Spin resonance studies of the quasi-one-dimensional Heisenberg antiferromagnet Cs_2CuCl_4

J.M. Schrama [a,*], A. Ardavan [a], A.V. Semeno [a], P.J. Gee [a], E. Rzepniewski [a], J. Suto [a], R. Coldea [a], J. Singleton [a], P. Goy [b]

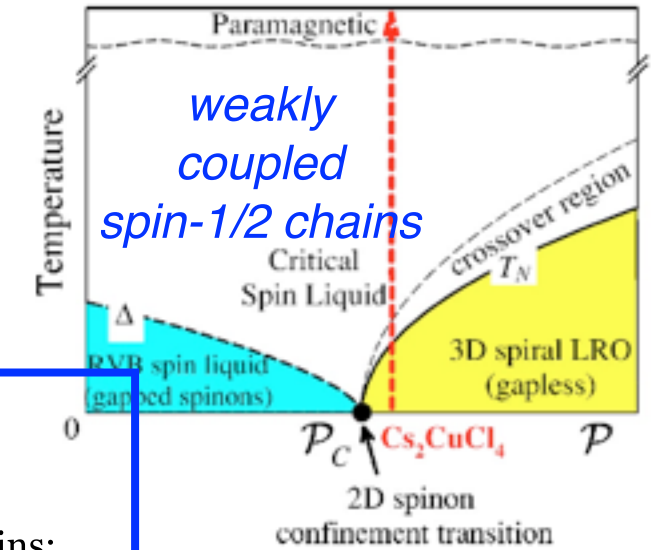
[a] *Clarendon Laboratory, University of Oxford, Parks Road, Oxford OX1 3PU, UK*
[b] *Abmm, 52 rue Lhomond, 75005 Paris, France*

H || c



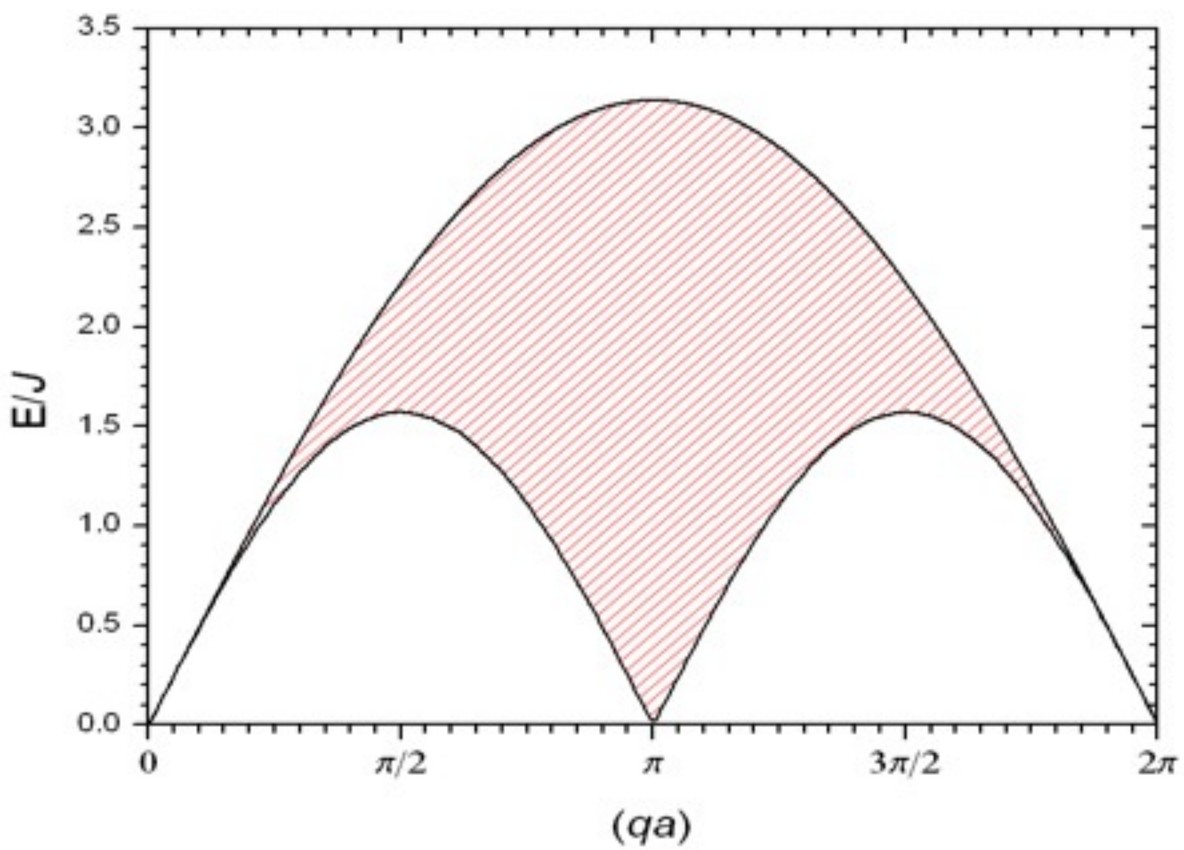
Temperature regimes

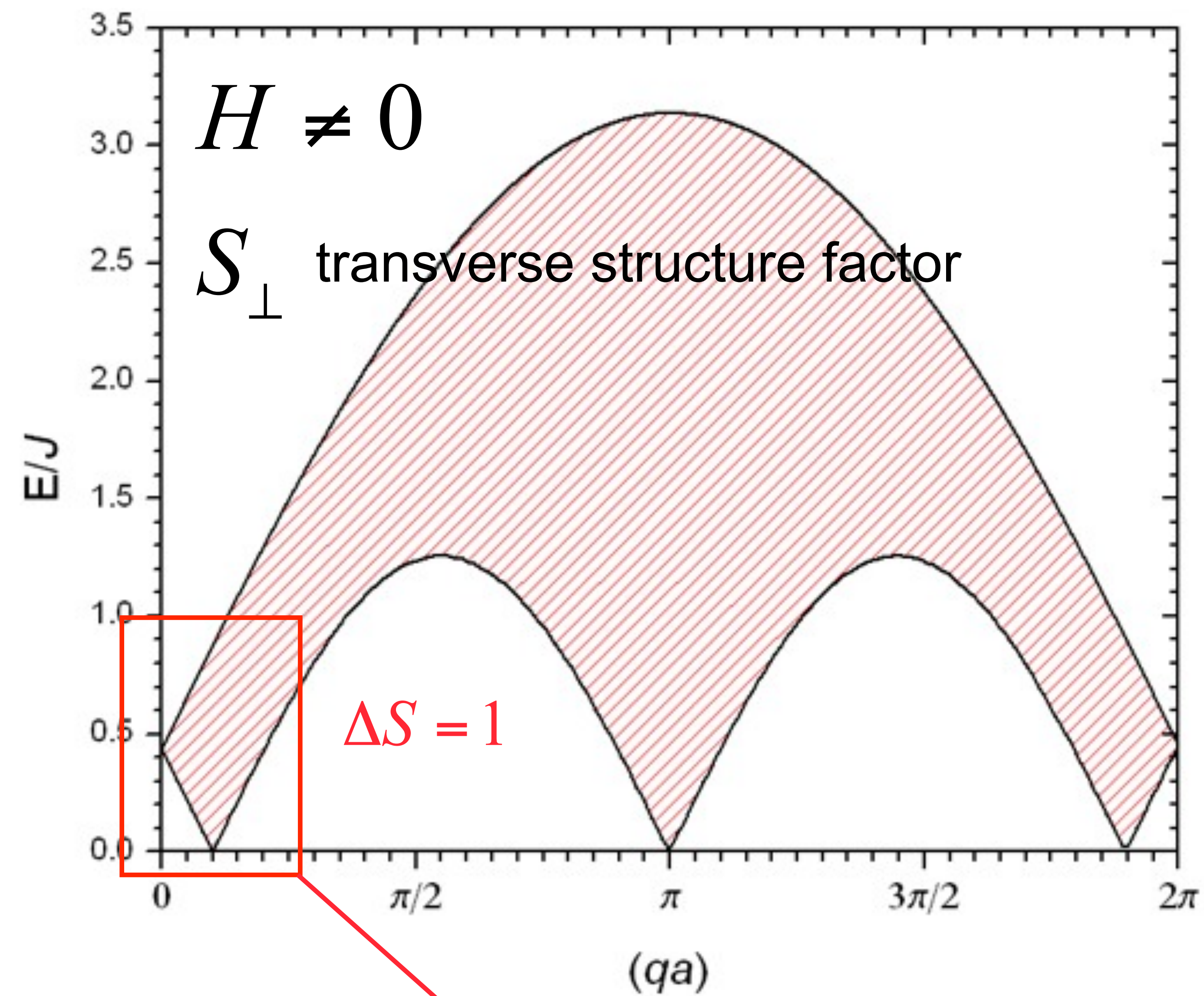
Paramagnetic individual spins	J S
universal quasi-classical regime	*correlated spins (high T field theory)*
spin-correlated (spin liquid) well-developed correlations along chains; but little correlations between chains	$T_0 \sim J\, e^{-2\pi S}$ Haldane scale does exist for S=1/2 chains: different response for S=1 and S=1/2 chains below this temperature *spinons* (low T field theory)
ordered phase strongly coupled chains; 2d (or 3d) description	$T_N = 0.6$ K spin waves (at low energy)

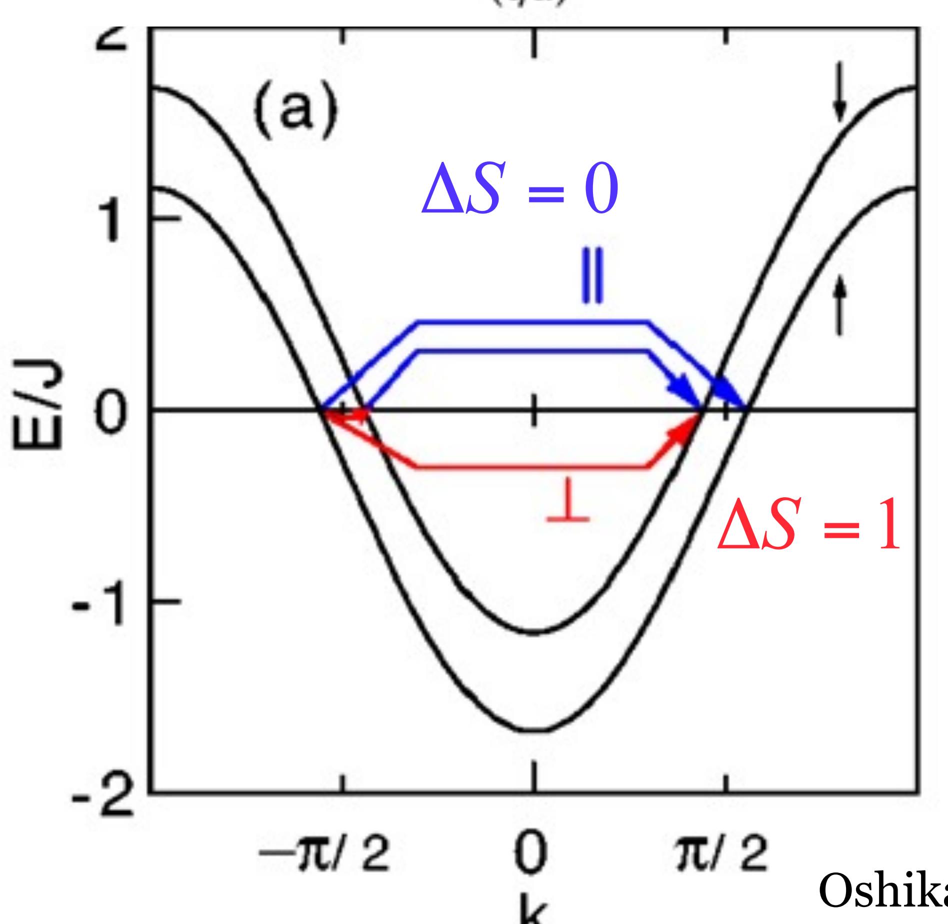


C. Buragohain, S. Sachdev
PRB 59 (1999)

Continuum in magnetic field

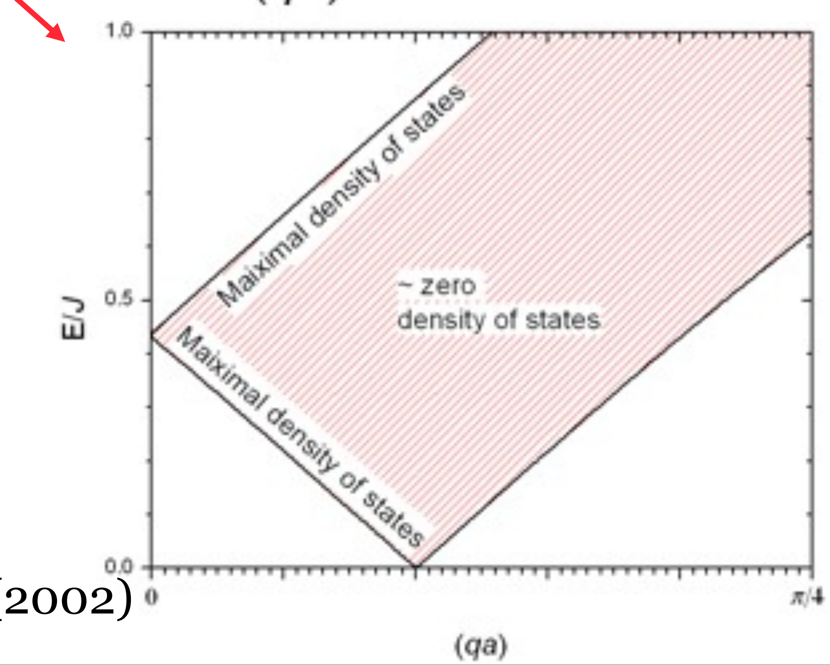






Dender et al, PRL 1997

Oshikawa, Affleck, PRB **65** 134410 (2002)



Explanation I: H along DM axis

$$H = \sum_{x,y,z} \boxed{J S_{x,y,z} \cdot S_{x+1,y,z}} - \boxed{D_{y,z} \cdot S_{x,y,z} \times S_{x+1,y,z}} - g\mu_B H \cdot S_{x,y,z}$$

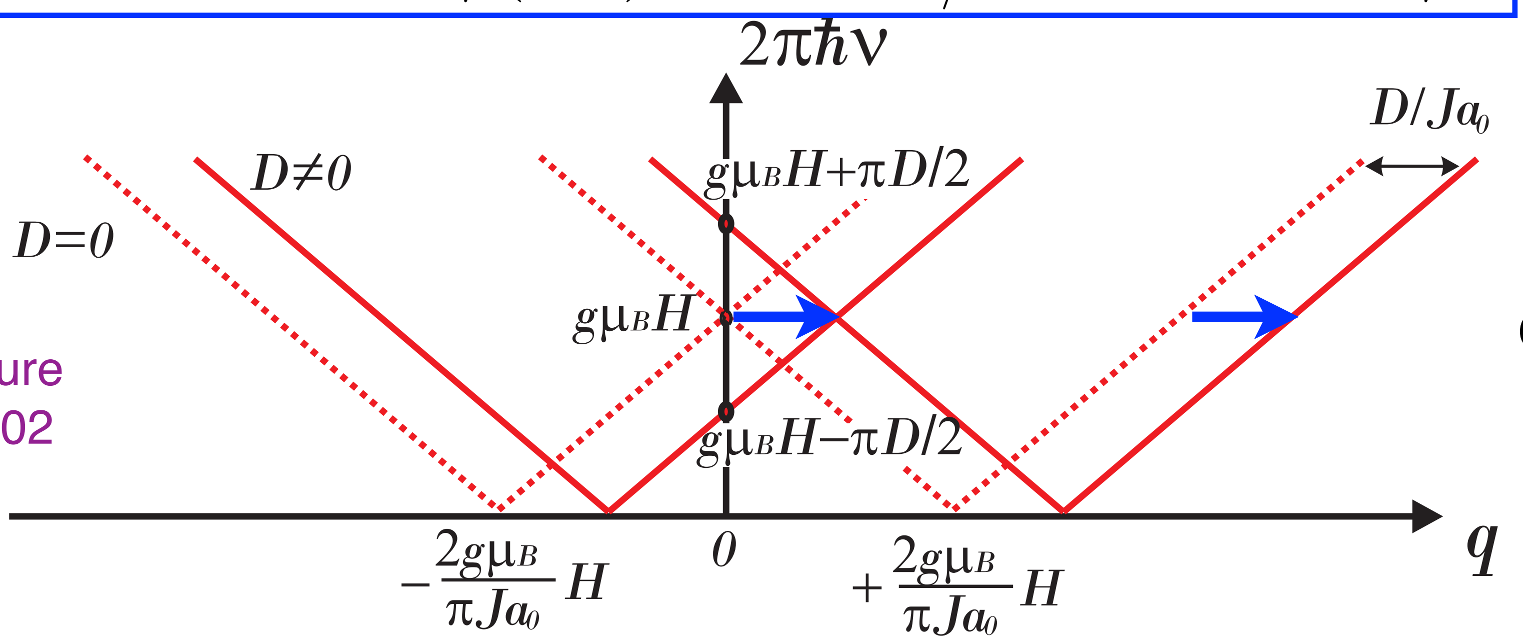
chain — uniform DM along the chain — magnetic field

!

Unitary rotation about **z**-axis $S^+(x) \to S^+(x) e^{i(D/J)x}, S^z(x) \to S^z(x)$

- removes DM term from the Hamiltonian (to D^2 accuracy)
- ***boosts*** momentum to $D/(J a_0)$

$$q = 0 \to q = D/(Ja_0) \Rightarrow 2\pi\hbar\nu_{R/L} = g\mu_B H \pm \pi D/2$$



dotted lines: D=0 picture
Oshikawa, Affleck 2002

rotated basis: q=0
original basis: q=D/J

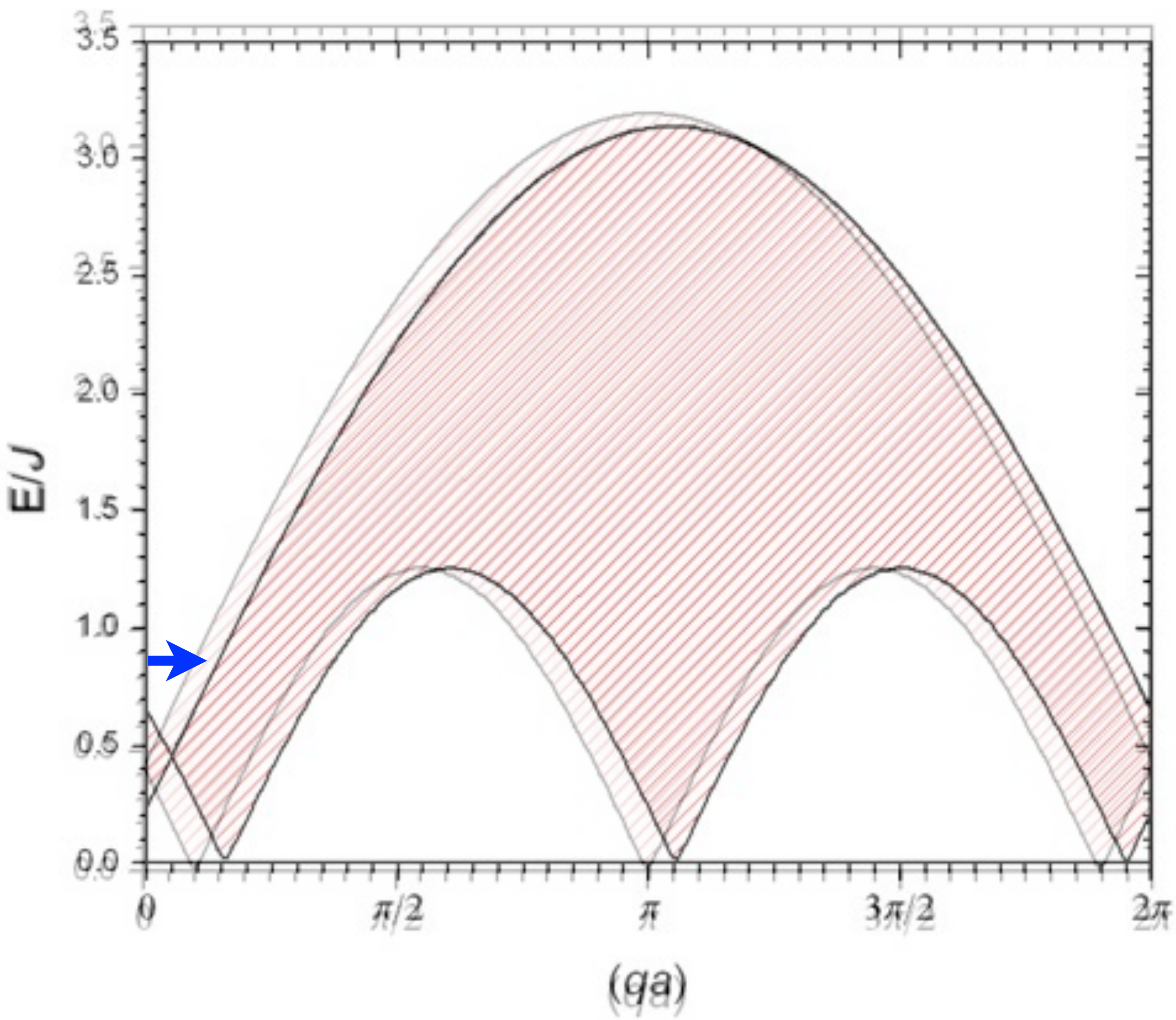
Chiral probe: **ESR probes *right*- and *left*-moving modes (spinons) independently**

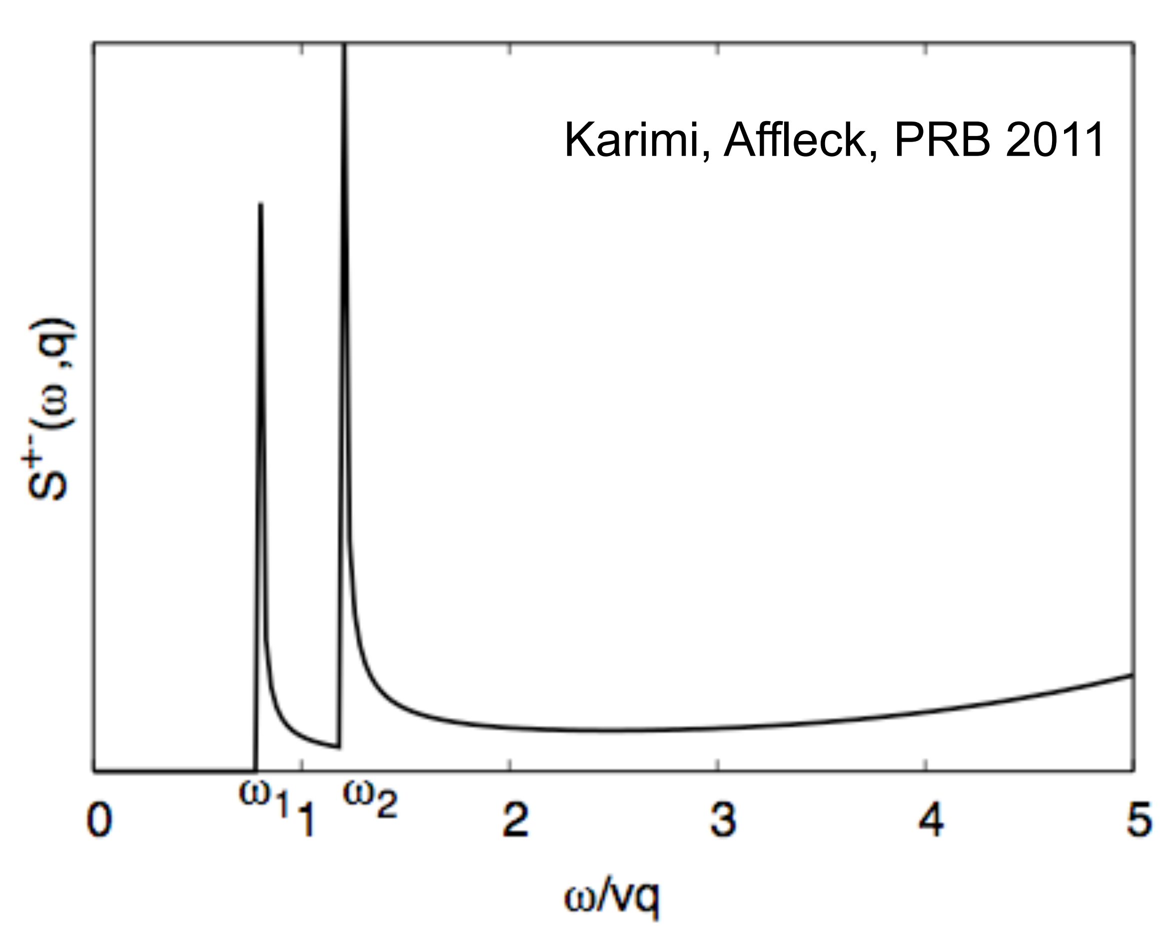
Spectrum shift due to the *uniform* DM

$$\mathcal{H} = \sum_n J(\mathbf{S}_n \cdot \mathbf{S}_{n+1}) + (\mathbf{D} \cdot [\mathbf{S}_n \times \mathbf{S}_{n+1}]) - \mu_B g(\mathbf{H} \cdot \mathbf{S}_n)$$

Gangadharaiah, Sun, Starykh, PRB **78** 054436 (2008)

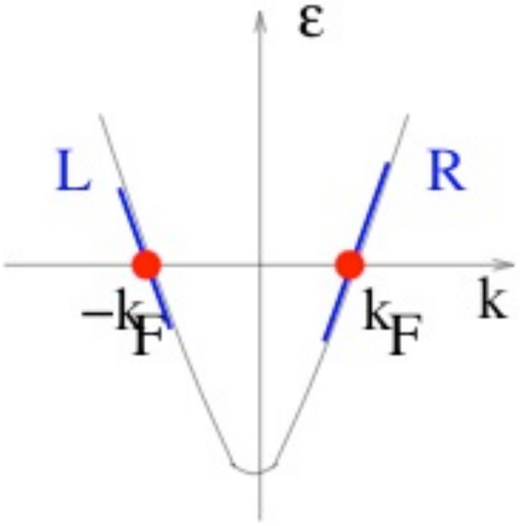
Spin transformation $S_n^+ = \widetilde{S}_n^+ e^{i\alpha n}$ $\alpha = -D/J$ **excludes DM interaction**, but **results in the spectrum shift** by momentum *D/J*. This leads to **two** ESR peaks.





Explanation II: arbitrary orientation

Relevant spin degrees of freedom



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

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

- *q=0 fluctuations: right (R) and left (L) spin currents*

$$\vec{M}_{R/L} = \Psi^\dagger_{R/L,s}\frac{\vec{\sigma}_{ss'}}{2}\Psi_{R/L,s'}$$

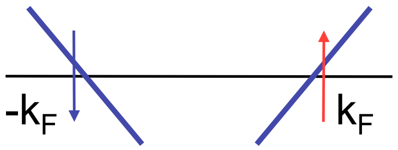
- *$2k_F$ (= π/a) fluctuations: charge density wave ε , spin density wave N*

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 $\chi_{1d}(q)$

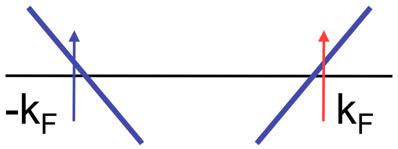
1/q

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



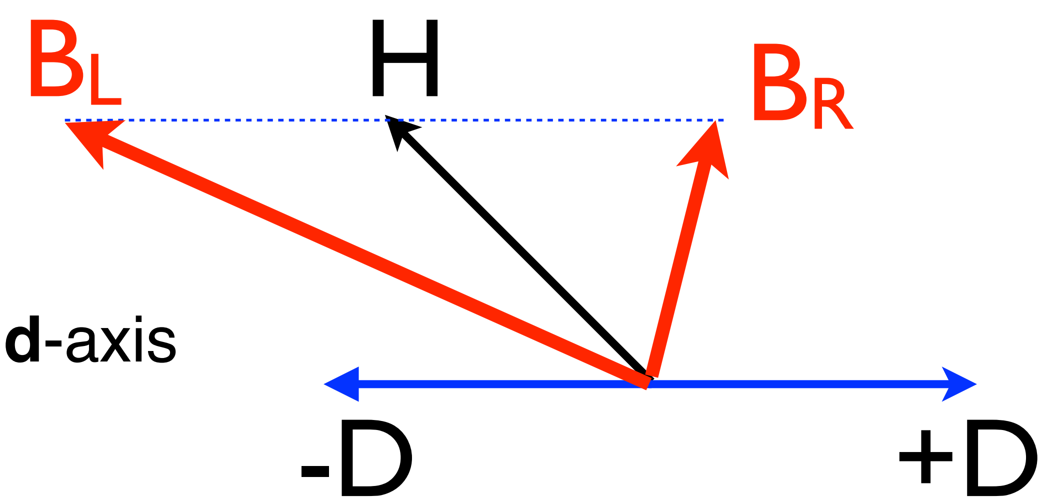
1/q

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

Explanation II: arbitrary orientation

$$H = \underbrace{\frac{2\pi v}{3}[(\vec{M}_R)^2 + (\vec{M}_L)^2]}_{\text{unperturbed chain}} \underbrace{- \frac{vD}{J}[M_R^d - M_L^d]}_{\text{uniform DM along the chain}} \underbrace{- g\mu_B H [M_R^z + M_L^z]}_{\text{magnetic field}}$$

unperturbed chain — uniform DM along the chain — magnetic field



Uniform DM produces internal momentum-dependent magnetic field along **d**-axis

- Total field acting on right/left movers $g\mu_B \vec{H} \pm \hbar v \vec{D}/J$
- Hence ESR signals at $2\pi\hbar\nu_{R/L} = |g\mu_B \vec{H} \pm \hbar v \vec{D}/J|$
- Polarization: for H=0 maximal absorption when microwave field h_{mw} is perpendicular to the internal (DM) one. Hence **h_{mw} II b** is most effective.

Povarov et al, PRL 2011
Gangadharaiah, Sun, OS, PRB 2008

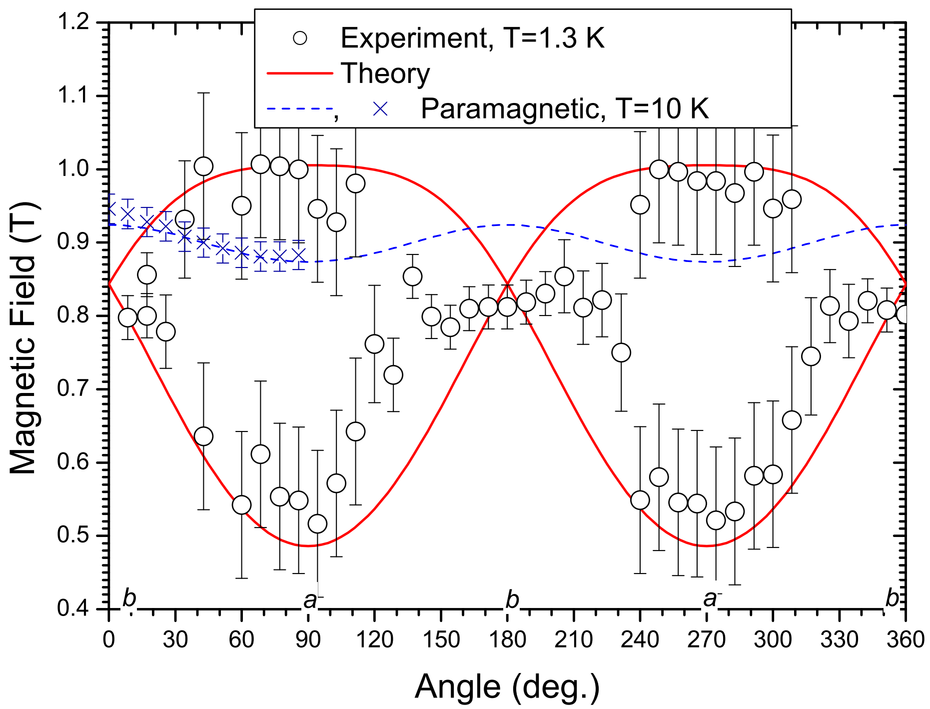
Explanation III: arbitrary orientation

- General orientation of **H** and **D**
- 4 sites/chains in unit cell

$$(2\pi\hbar\nu_R)^2 = (g_b\mu_B H_b)^2 + [g_a\mu_B H_a + (-1)^z \pi D_a/2]^2 + [g_c\mu_B H_c + (-1)^y \pi D_c/2]^2,$$

$$(2\pi\hbar\nu_L)^2 = (g_b\mu_B H_b)^2 + [g_a\mu_B H_a - (-1)^z \pi D_a/2]^2 + [g_c\mu_B H_c - (-1)^y \pi D_c/2]^2.$$

a-b plane

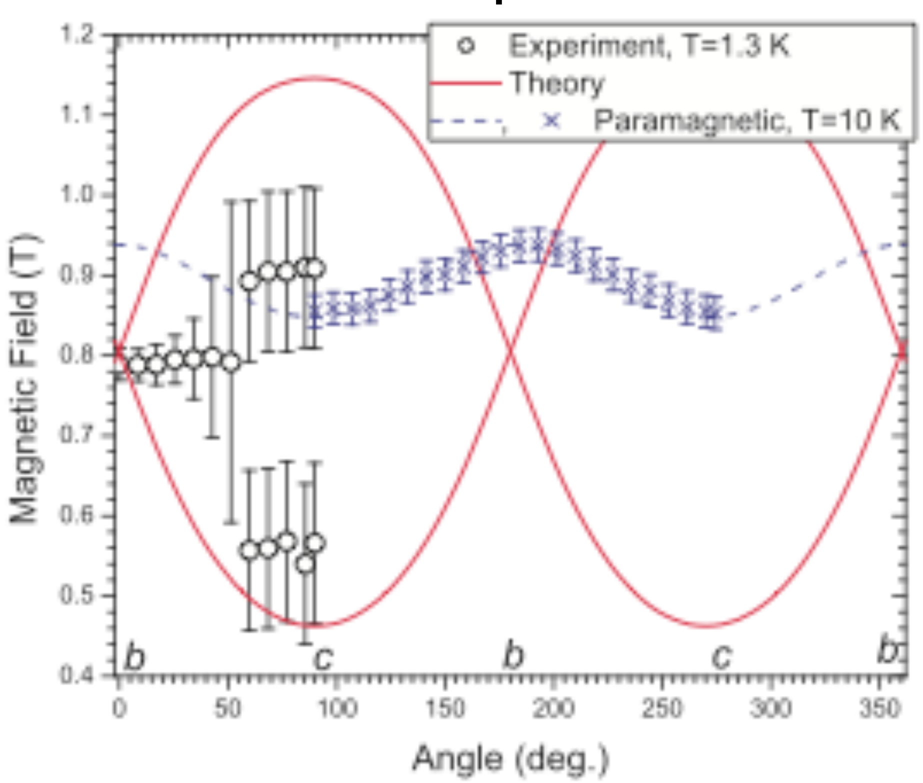


$$D_a/(4\hbar) = 8\ \text{GHz}$$
$$D_c/(4\hbar) = 11\ \text{GHz}$$

0.3 Tesla
0.4 Tesla

D ~ J/10

b-c plane



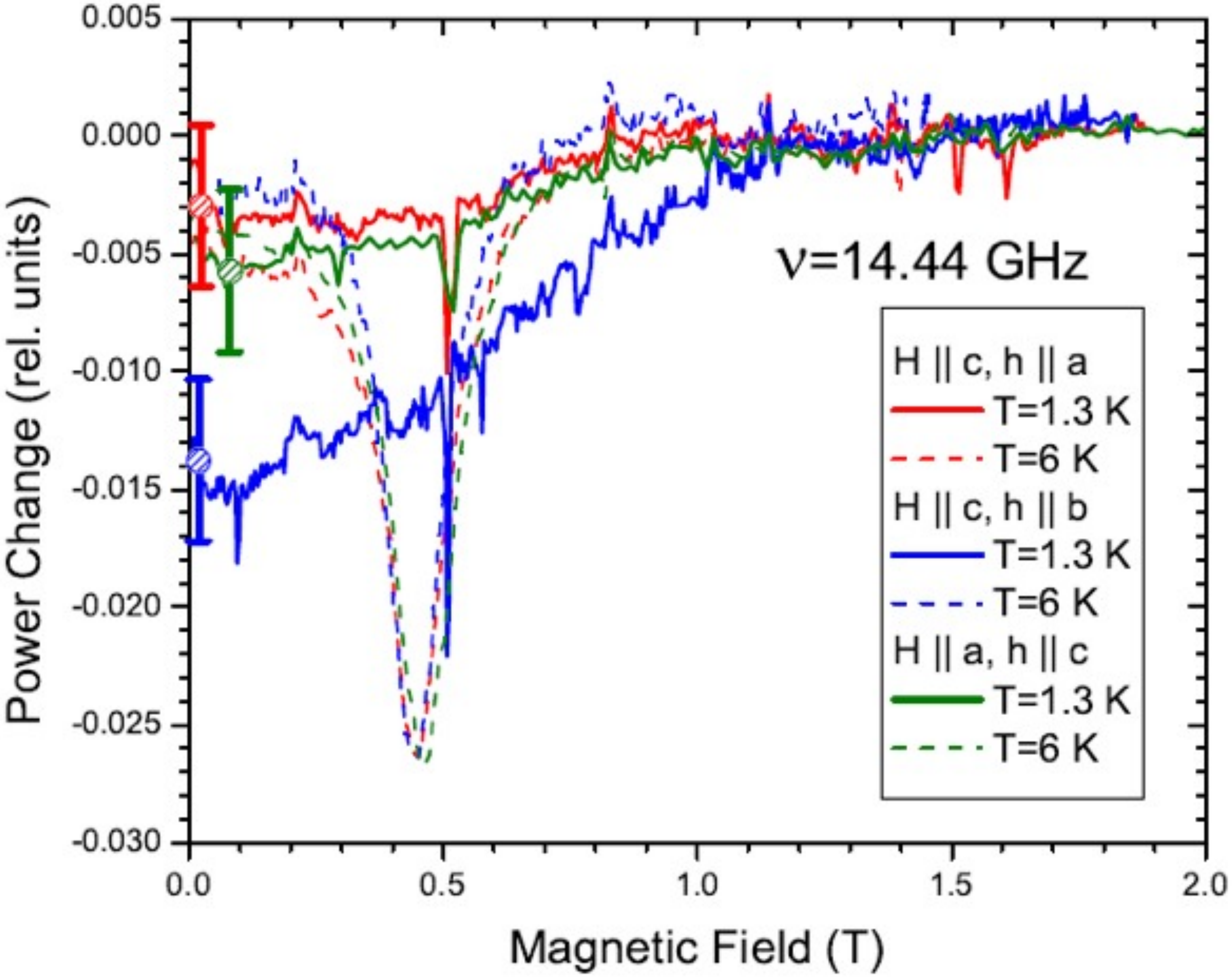
- for **H** along b-axis only: the "gap" is determined by the DM interaction strength

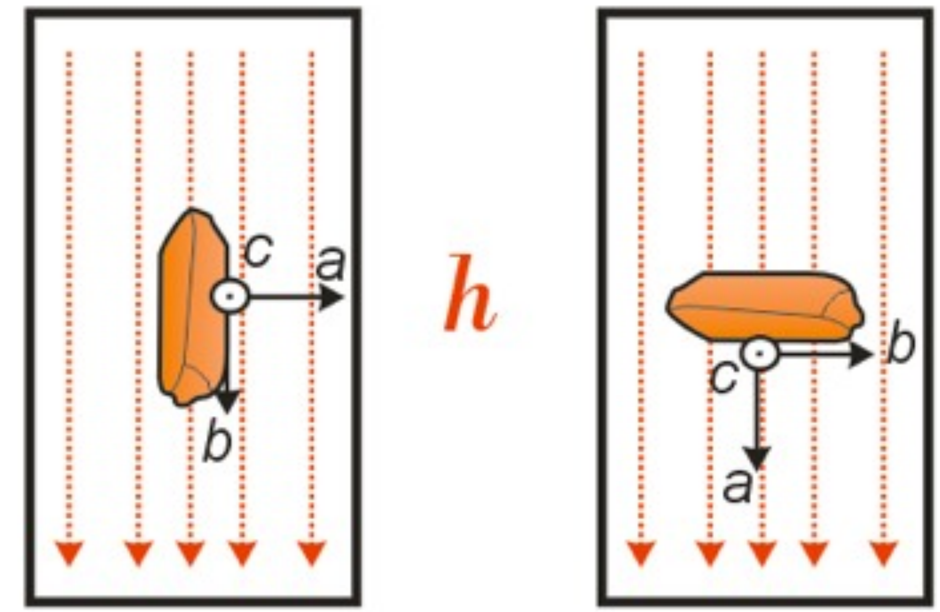
$$\Delta = \frac{\pi}{2}\sqrt{D_a^2 + D_c^2} \rightarrow (2\pi\hbar)13.6\ \text{GHz}$$

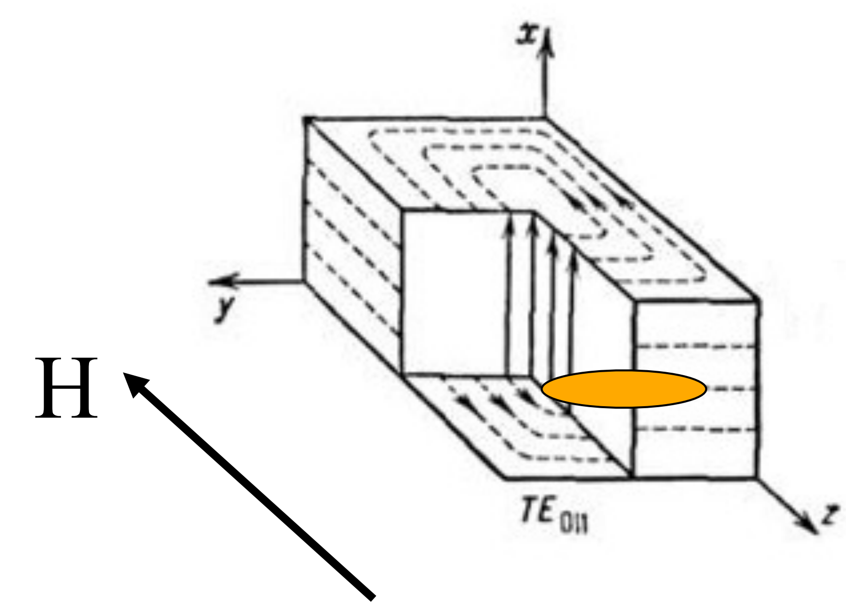
This explanation suggests:

1) ESR absorption in the absence of H $\nu \sim \sqrt{D_a^2 + D_c^2}$

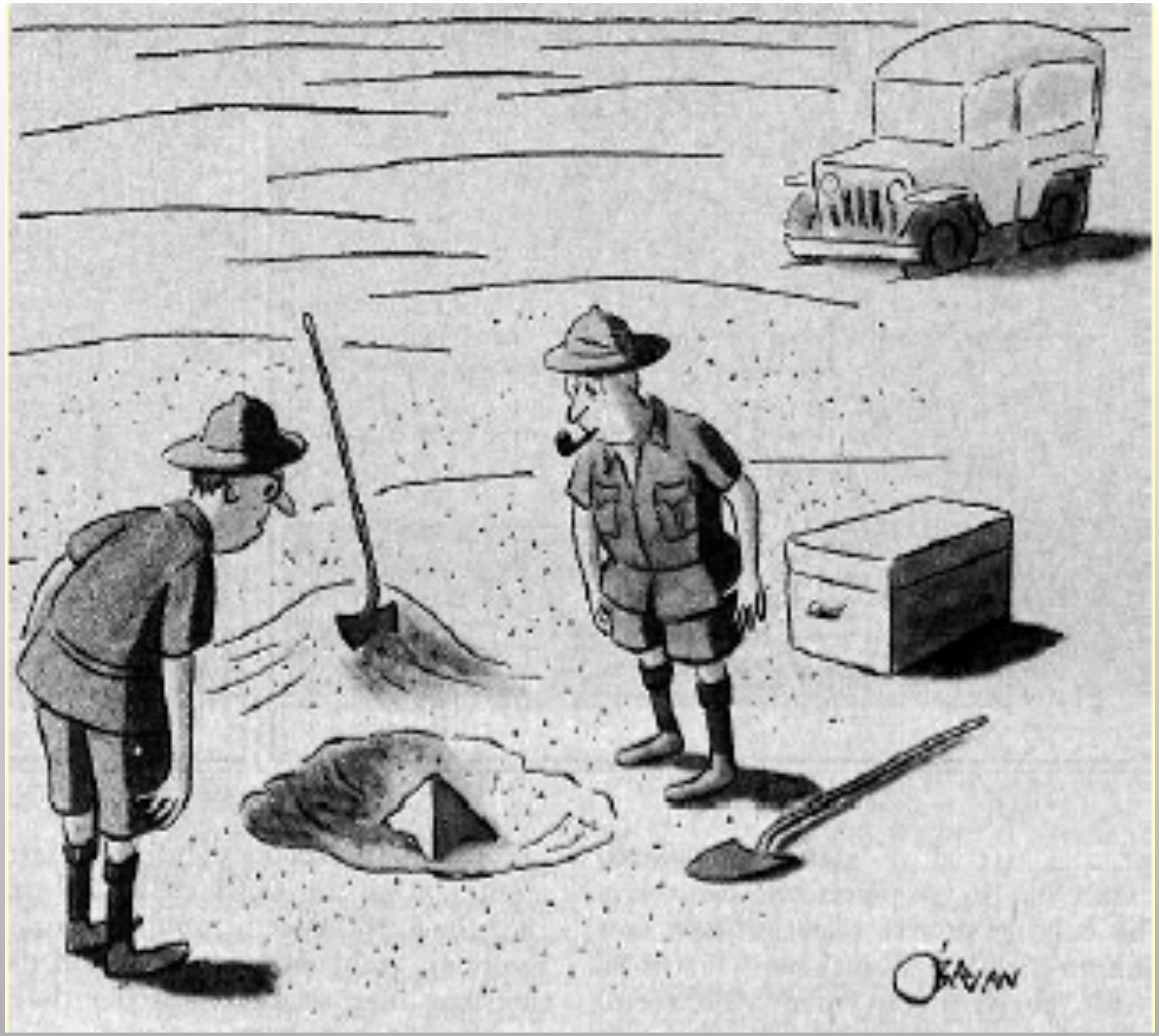
2) strong polarization dependence in zero field







The largest absorption occurs when microwave field h(t) is lined along crystal b-axis, h || *b* [so that it is perpendicular to the **D** vector in *a*-*c* plane]



“This could be the discovery of the century. Depending, of course, on how far down it goes”

Extension to two-dimensional spin liquids with spinon Fermi surface

Higher dimensional extension (weak Mott insulators)

- origin of DM: spin-orbit tunneling in Hubbard model

$$\hat{H} = \sum_{i,j} \{c^{+}_{i,\alpha}(-t\delta_{\alpha\beta} + i\vec{\lambda}_{ij} \cdot \vec{\hat{s}}_{\alpha\beta})c_{j,\beta} + \text{H.c.}\} + U\sum_{i} n_{i\uparrow}n_{i\downarrow}.$$

- 2D square lattice with ***uniform spin-orbit*** interaction ($YBa_2\ Cu_3\ O_{6+x}$)

$$\vec{\lambda}_{ij} = \lambda\hat{z} \times (\vec{r}_i - \vec{r}_j)$$

Coffey, Rice, Zhang 1991
Shekhtman, Entin-Wolhman, Aharony 1992
Bonesteel 1993

- (Lattice) spin-orbit interaction of Rashba type

$$\hat{H}_{\text{SO}}(\mathbf{k}) = -2\lambda \sum_{k} c^{\dagger}_{k,\alpha}\{\hat{s}_x \sin[k_y] - \hat{s}_y \sin[k_x]\}c_{k,\beta}$$

- Transition to ***spinons*** via ***slave-rotor*** formulation $c_{r,\sigma} = f_{r,\sigma}e^{i\theta_r}$

Florens and Georges 2004
S.-S. Lee and P. A. Lee 2005

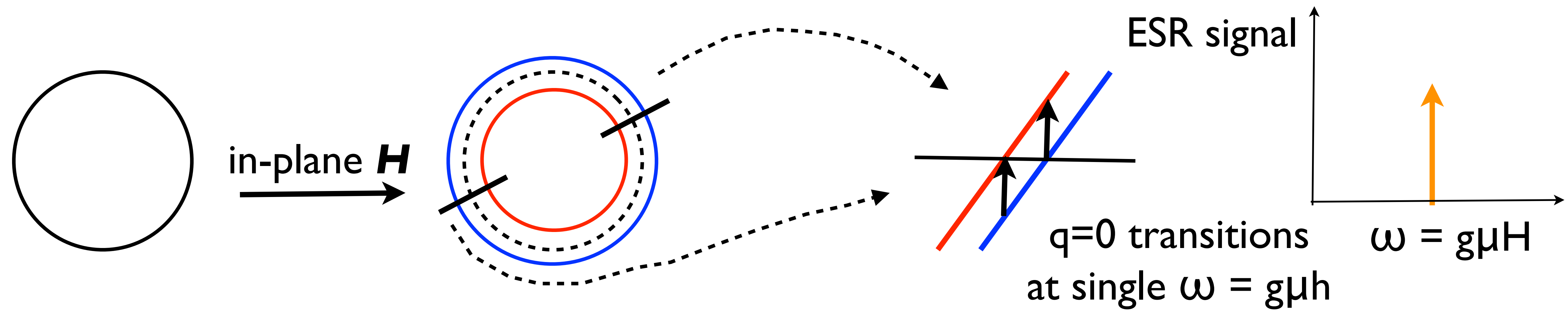
- (mean-field) Rashba Hamiltonian for free spinons ($f_{r,s}$)

$$\hat{H}_f = \sum_{i,j} f^{+}_{i,\alpha}(-t\delta_{\alpha\beta} + i\vec{\lambda}^{\text{eff}}_{ij} \cdot \vec{s}_{\alpha\beta})f_{j,\beta} - \vec{H} \cdot f^{+}_{i,\alpha}\vec{s}_{\alpha\beta}f_{j,\beta}$$

Glenn, OS, Raikh, PRB 2012

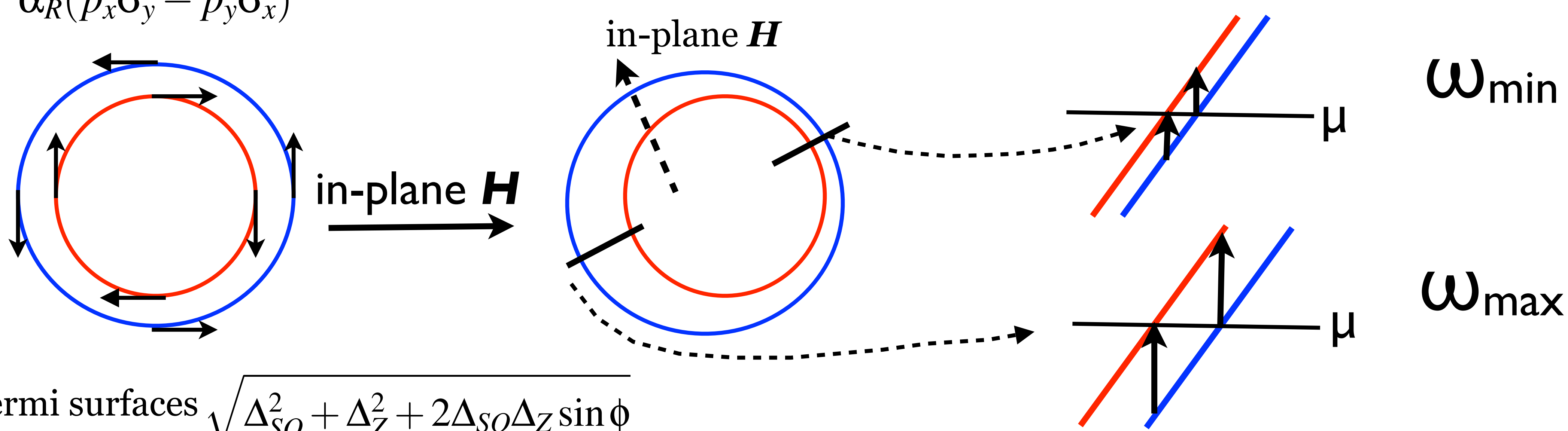
Estimates for 2d spinon gas using Rashba model as an example

no DM



DM

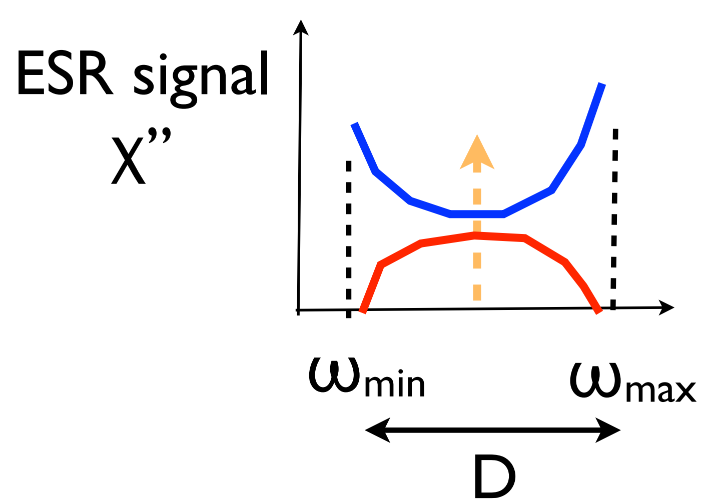
$\alpha_R(p_x\sigma_y - p_y\sigma_x)$



splitting of Fermi surfaces $\sqrt{\Delta_{SO}^2 + \Delta_Z^2 + 2\Delta_{SO}\Delta_Z \sin\phi}$

Raikh, Chen 1999

Energy absorption due to microwave h(t)



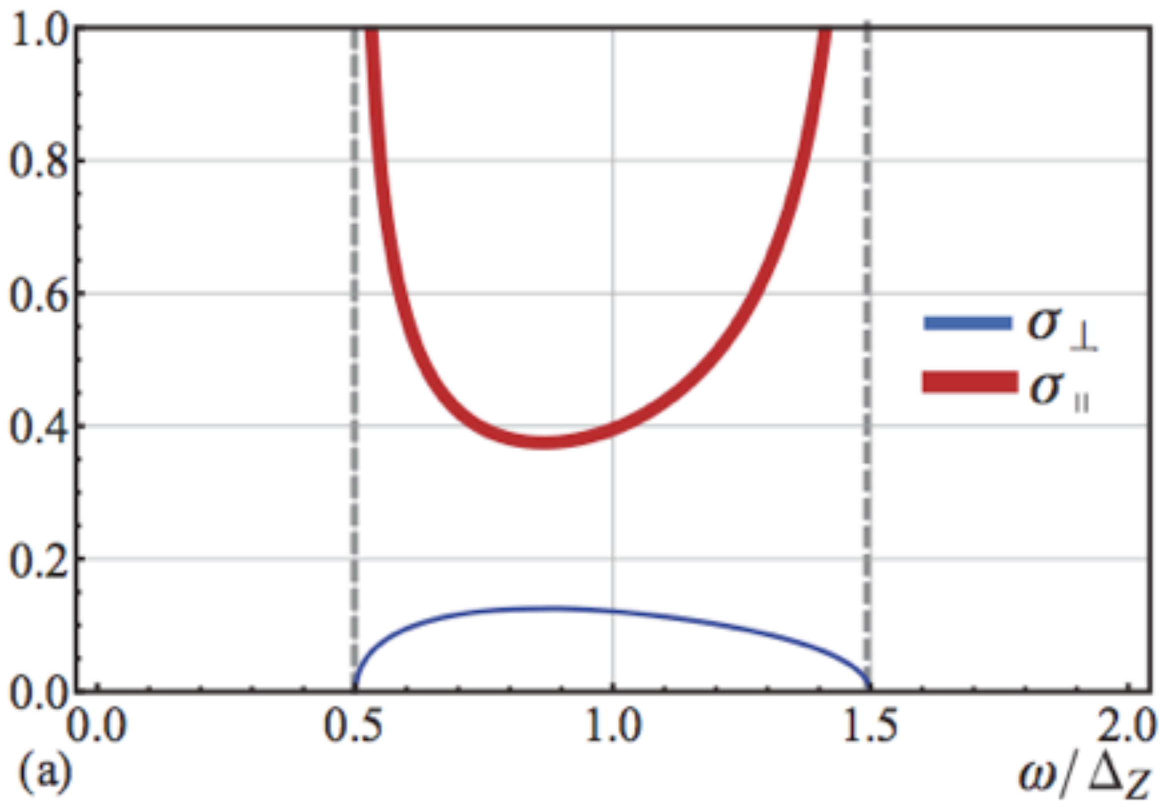
the width of the line ~ min(H, α_R k_F)

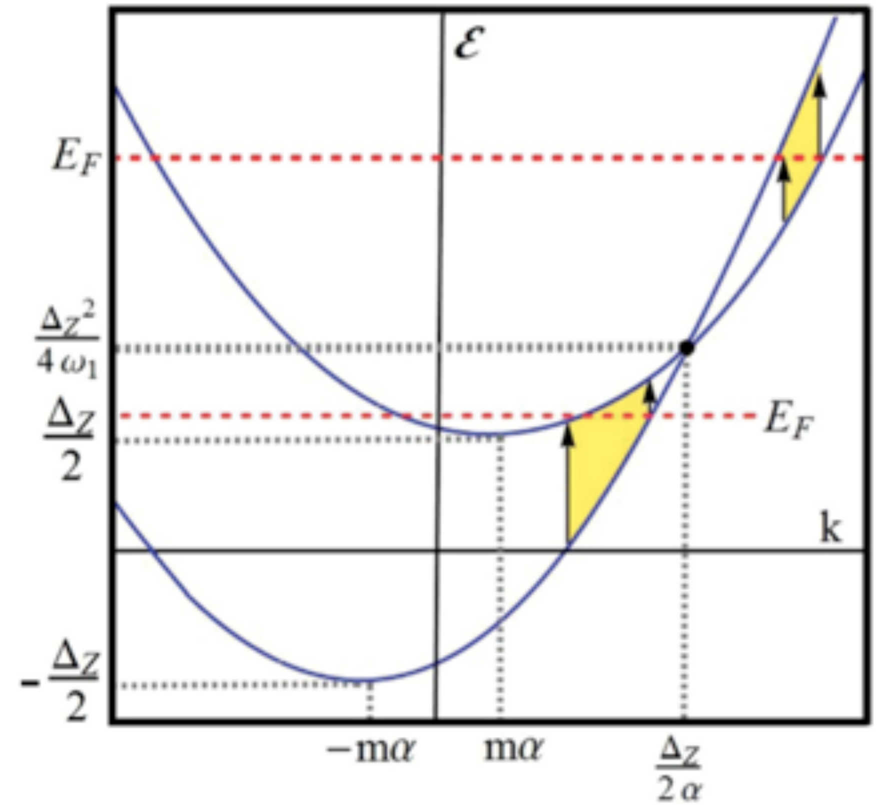
line shape is strongly polarization-dependent:

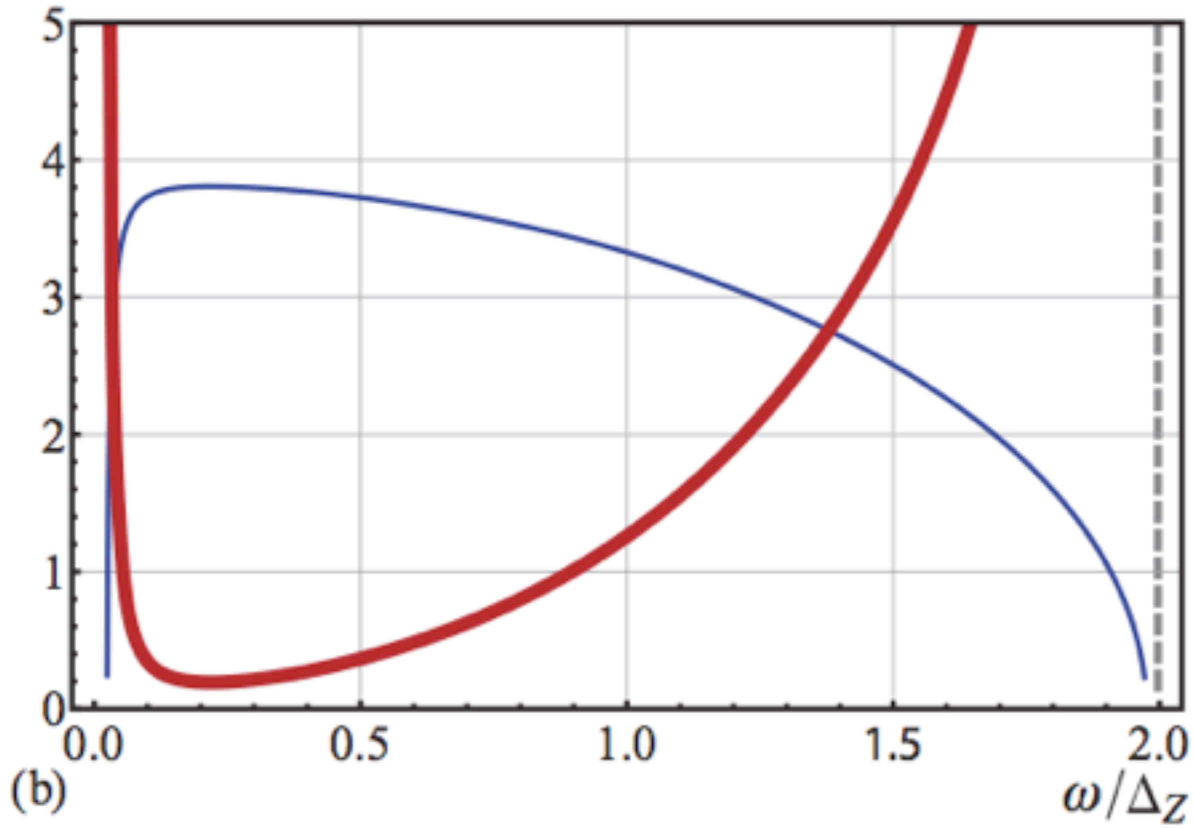
$\left[\omega - \omega_{min/max}\right]^{-1/2}$ for h(t) perpendicular to H

$\left[\omega - \omega_{min/max}\right]^{1/2}$ for h(t) parallel to H

Non-trivial lineshape due to vertical transitions between asymmetric subbands







the width of the line ~ min(H, α_R k_F)

line shape is strongly polarization-dependent:

$[\omega - \omega_{min/max}]^{-1/2}$ for h(t) perpendicular to H

$[\omega - \omega_{min/max}]^{1/2}$ for h(t) parallel to H

$$\chi'' \sim \omega\sigma(\omega)$$

Glenn, OS, Raikh PRB 2012

Conclusions

- DM can be used to probe exotic spin liquids
- 1D: ESR is a chiral probe of critical spinons (neutral fermions)
  - measurements at small momentum ~ D/J
  - allows to extract parameters of DM (spin-orbit) interaction
- Higher-dimensional extension: DM breaks SU(2) and provides access to spinon Fermi surface

Another geometry: **staggered** DM

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Direct Observation of Field-Induced Incommensurate Fluctuations in a One-Dimensional $S = 1/2$ Antiferromagnet

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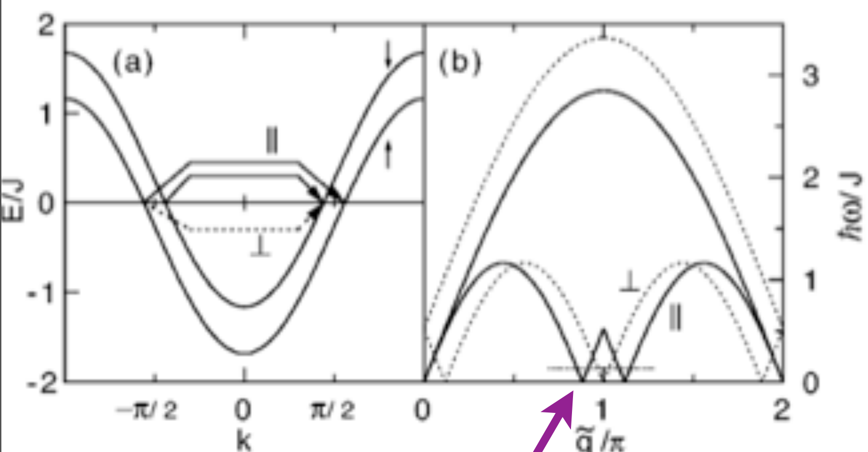
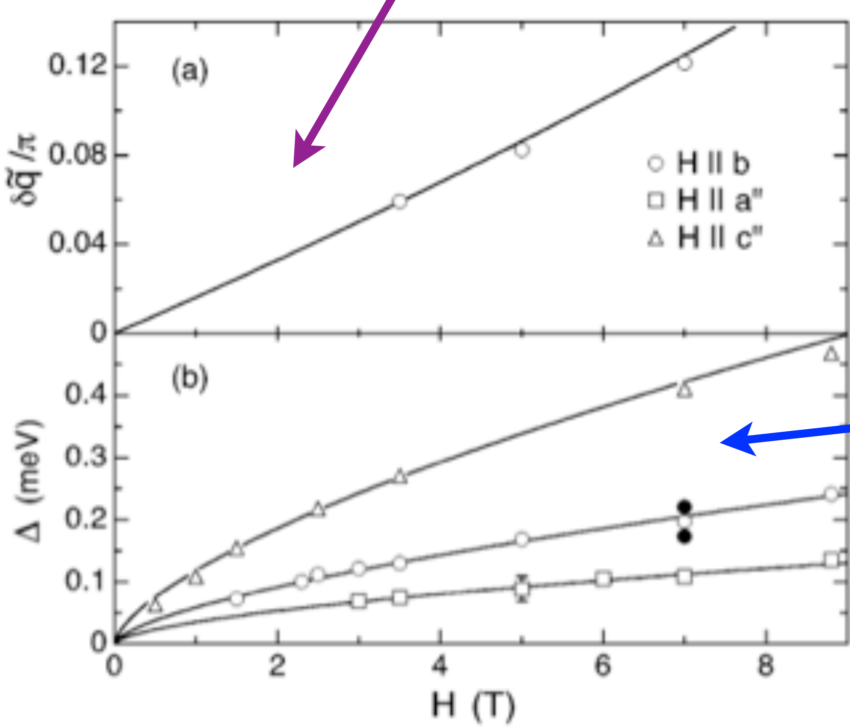
(Received 31 March 1997)

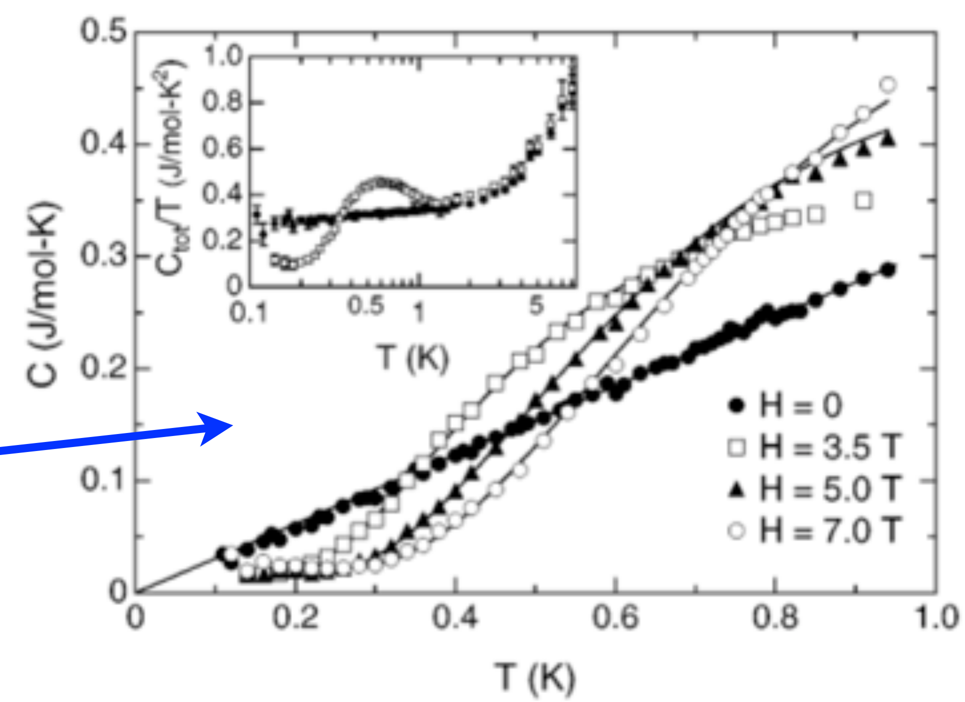
Neutron scattering from copper benzoate, $Cu(C_6D_5COO)_2 \cdot 3D_2O$, provides the first direct experimental evidence for field-dependent incommensurate low energy modes in a one-dimensional spin $S = 1/2$ antiferromagnet. Soft modes occur for wave vectors $\tilde{q} = \pi \pm \delta\tilde{q}(H)$, where $\delta\tilde{q}(H) \approx 2\pi M(H)/g\mu_B$ as predicted by Bethe ansatz and spinon descriptions of the $S = 1/2$ chain. Unexpected was a field-induced energy gap $\Delta(H) \propto H^{\alpha}$, where $\alpha = 0.65(3)$ as determined from specific heat measurements. At $H = 7$ T ($g\mu_B H/J = 0.52$), the magnitude of the gap varies from $0.06J$ to $0.3J$ depending on the orientation of the applied field. [S0031-9007(97)03950-1]





field-induced shift

field-induced gap!



Oshikawa, Affleck
Essler, Tsvelik

$$\sum_x (-1)^x D \cdot S_x \times S_{x+1}$$

but: single ESR line!

Uniform *vs* staggered DM

$\vec{D}\cdot\vec{S}_n\times\vec{S}_{n+1}$

h=0: free spinons

finite h: free spinons

but *subject to momentum-dependent magnetic field*

ESR: generically two lines

- ✓ splitting
- ✓ shift
- ✓ width (?)

$(-1)^n\vec{D}\cdot\vec{S}_n\times\vec{S}_{n+1}$

free spinons

confined spinons

generate strongly relevant *transverse magnetic field that binds spinons together* $(-1)^n\frac{\vec{D}\times\vec{h}}{2J}\cdot\vec{S}_n$

single line

- ✓ shift
- ✓ width