Spinon magnetic resonance

Oleg Starykh, University of Utah



BigMag@UCSB

A workshop to identify transformational science that would be enabled by coupling a ~30 T magnet to UCSB's Terahertz Free-Electron Lasers

May 17-19, 2018

IOP Publishing

Journal of Physics: Condensed

Topical Review

Electrodynamics of quantum spin liquids

Martin Dressel and Andrej Pustogow

1. Physikalisches Institut, Universität Stuttgart, Pfaffenwaldring 57, 70550 Stuttgart, Germany

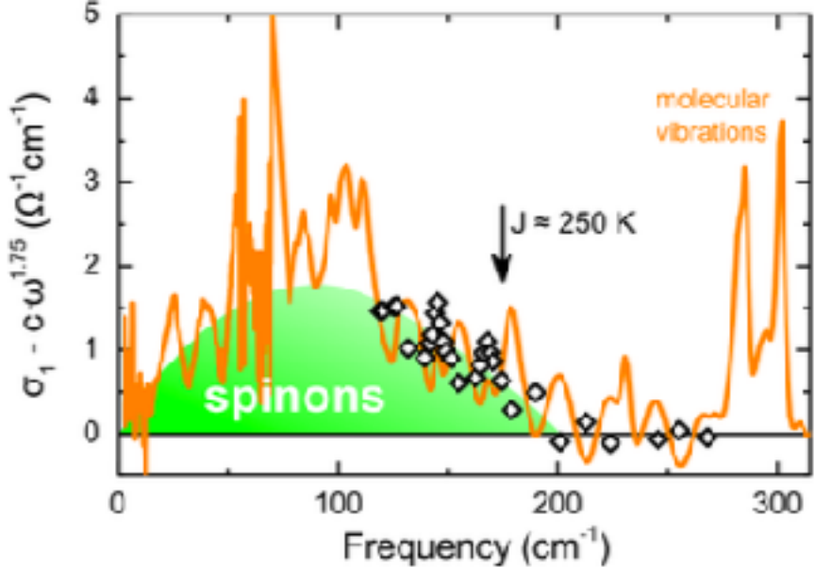


Figure 13. After subtracting the power-law background related with the Mott–Hubbard band, we obtain a broad low-energy feature below approximately 200cm^{-1}, which is close to the antiferromagnetic exchange energy of β'-EtMe$_3$Sb[Pd(dmit)$_2$]$_2$. Hence, we associate it with the coherent spinon Fermi surface predicted previously [60, 62]. The dome-like shape of the spinon contribution arises due to the rapid decay towards $\omega \to 0$, consistent with a power law such as the postulated ω^2 behavior [62]. This feature becomes visible when the background conductivity is sufficiently suppressed due to opening of the Mott gap. The strong, narrow modes at higher frequencies correspond to vibrational features.

Examples of current literature

200 cm^{-1} = 6 THz

PRL 119, 227201 (2017) PHYSICAL REVIEW LETTERS week ending 1 DECEMBER 2017

Antiferromagnetic Resonance and Terahertz Continuum in α-RuCl$_3$

A. Little,[1,2] Liang Wu,[1,2,3,*] P. Lampen-Kelley,[4,5] A. Banerjee,[5] S. Patankar,[1,2] D. Rees,[1,2] C. A. Bridges,[7] J.-Q. Yan,[8] D. Mandrus,[4,5] S. E. Nagler,[6,9] and J. Orenstein[1,2]

Spinons?

4 meV = 1 THz

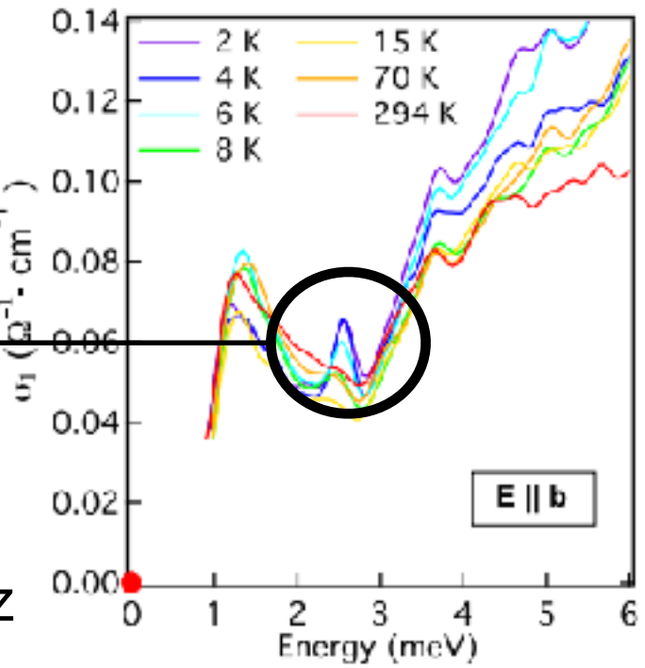
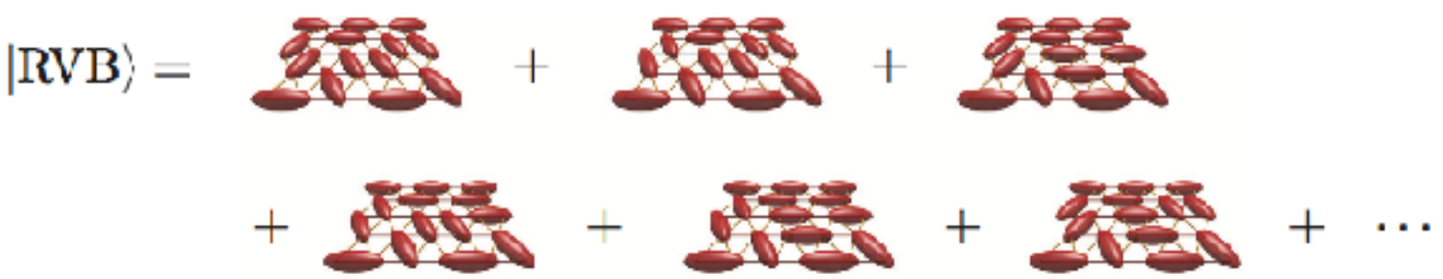


FIG. 3. Absorption spectra interpreted as optical conductivity, with **E** parallel to **b**.

The big question(s)

What is quantum spin liquid?



No broken symmetries.
Quantum entangled state:
fractionalized excitations = spinons
emergent gauge fields

Figure 1. A 'resonating valence bond' (RVB) state. Ellipsoids indicate spin-zero singlet states of two $S = 1/2$ spins.

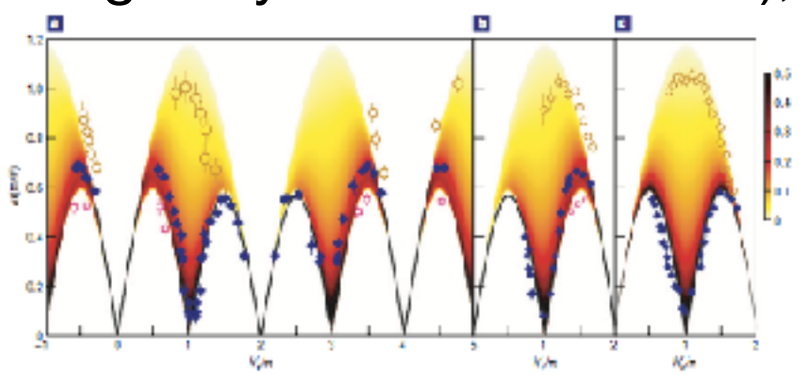
Savary, Balents 2017

Which materials realize it?

Past candidates: Cs2CuCl4, kagome volborthite…
Current candidates: kagome herbertsmithite, α-RuCl3, organic Mott insulators

How to detect/observe it?

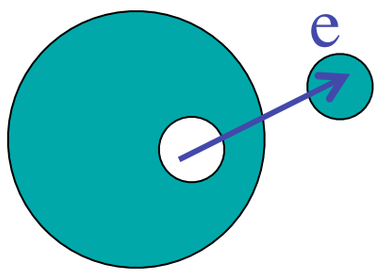
Neutrons (if good single crystals are available), NMR, ESR



Excitations: quasi-particles and their fractions

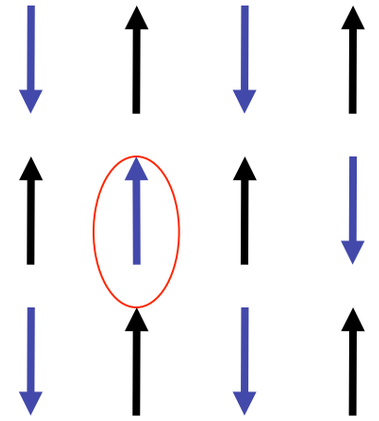
Conventional matter: quasi-particles = (renormalized) particles of underlying microscopic system

Fermi-liquid



Charge e, spin 1/2
but renormalized mass m^*

Neel state

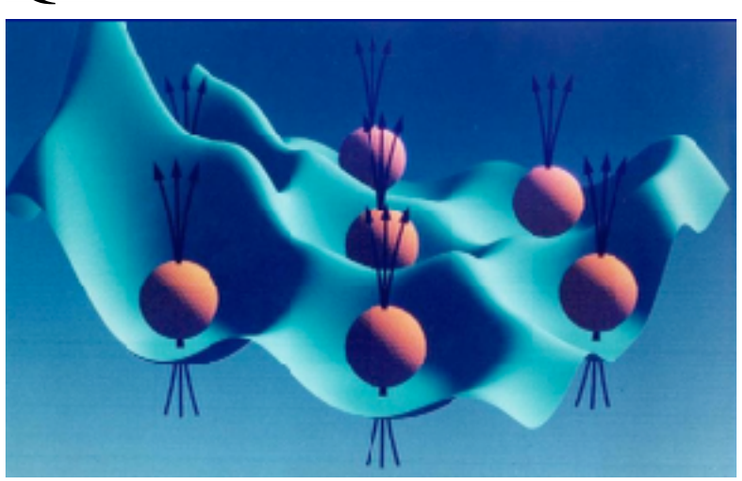


Spin-wave

Spin S=1
Adiabatically connected to spin-flip excitation of a single-spin in magnetic field

Strongly correlated matter: excitations carry a fraction of particle's quantum number

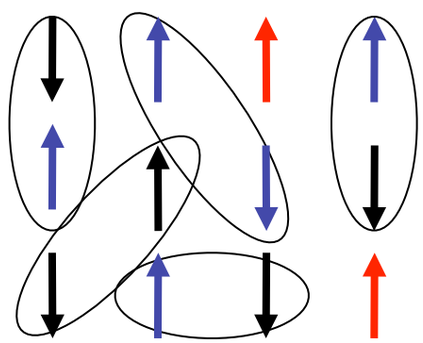
Quantum Hall effect



Tom Duff, Lucent

Charge e/3

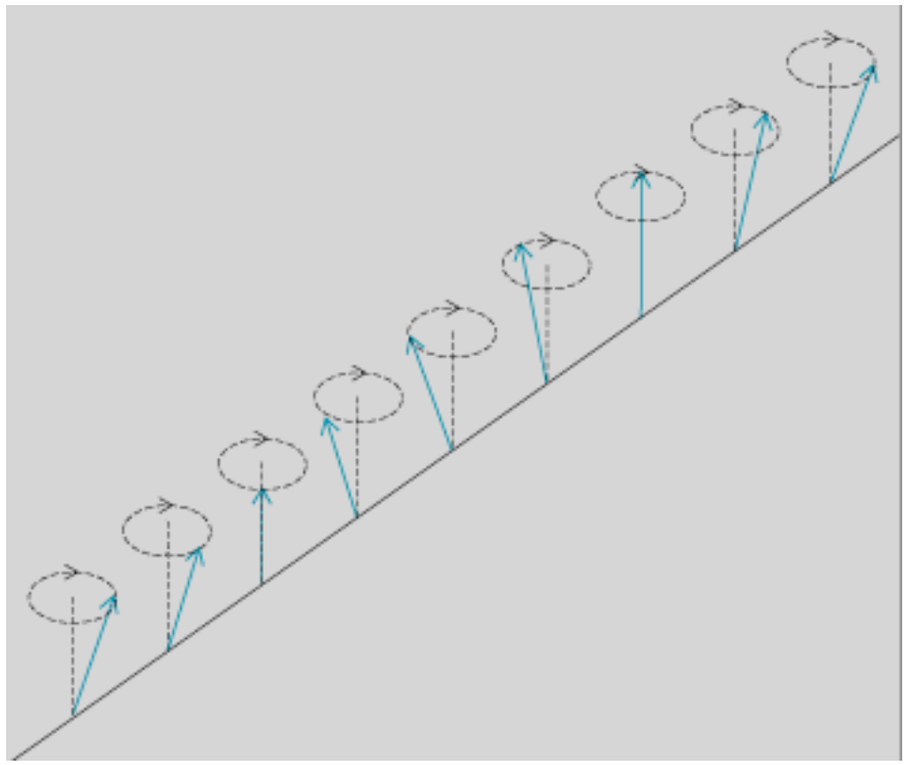
Spin liquid



Excitation -
broken valence bond:
two S=1/2 spinons

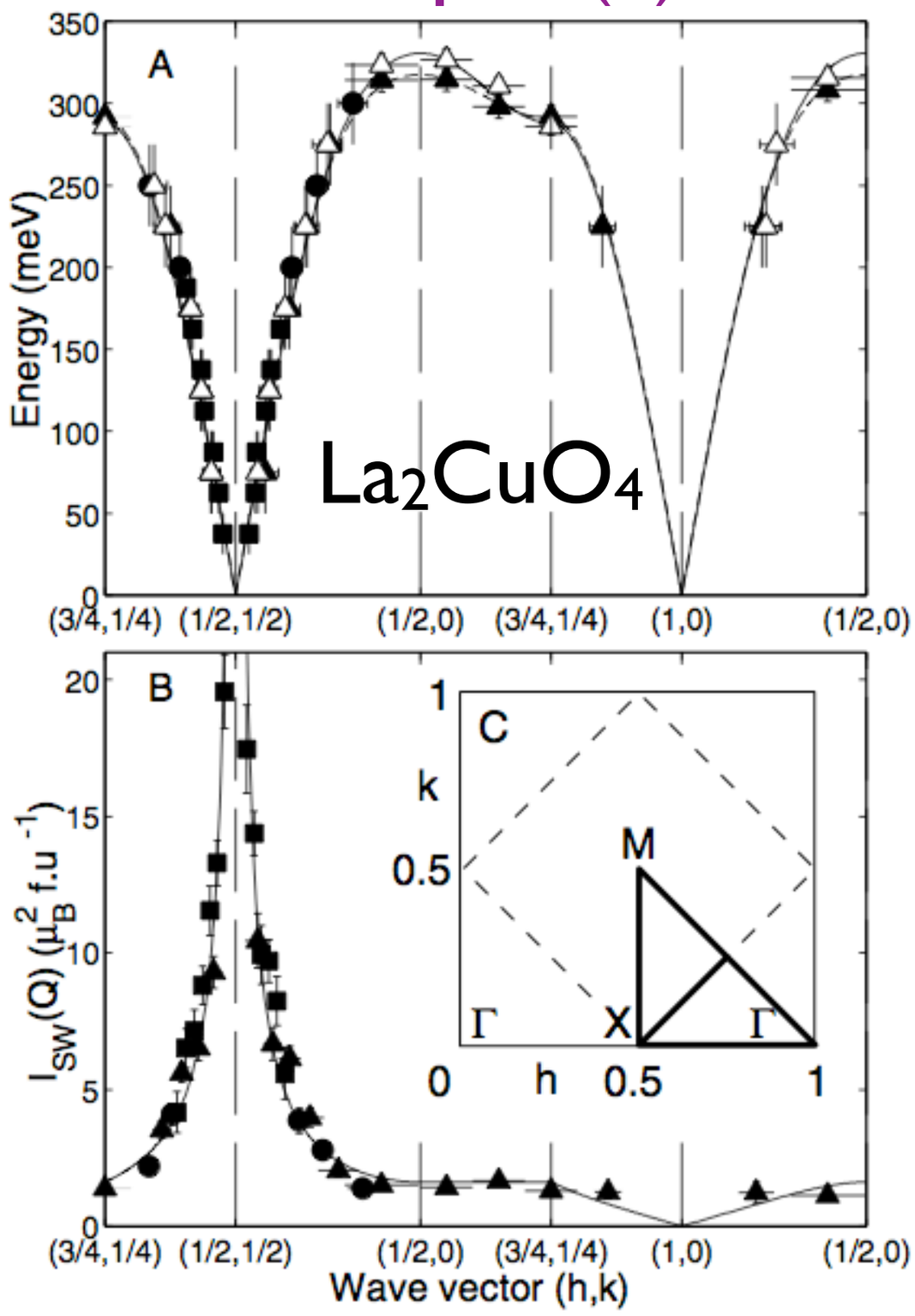
?

Spin wave or magnon = propagating disturbance in magnetically ordered state (ferromagnet, **antiferromagnet**, ferrimagnet...)



$\Sigma_r S^+_r e^{i\, k.r} |0\rangle$

sharp $\omega(k)$



Carries $S^z = 1$.
Observed via inelastic neutron scattering as a sharp single-particle excitation.

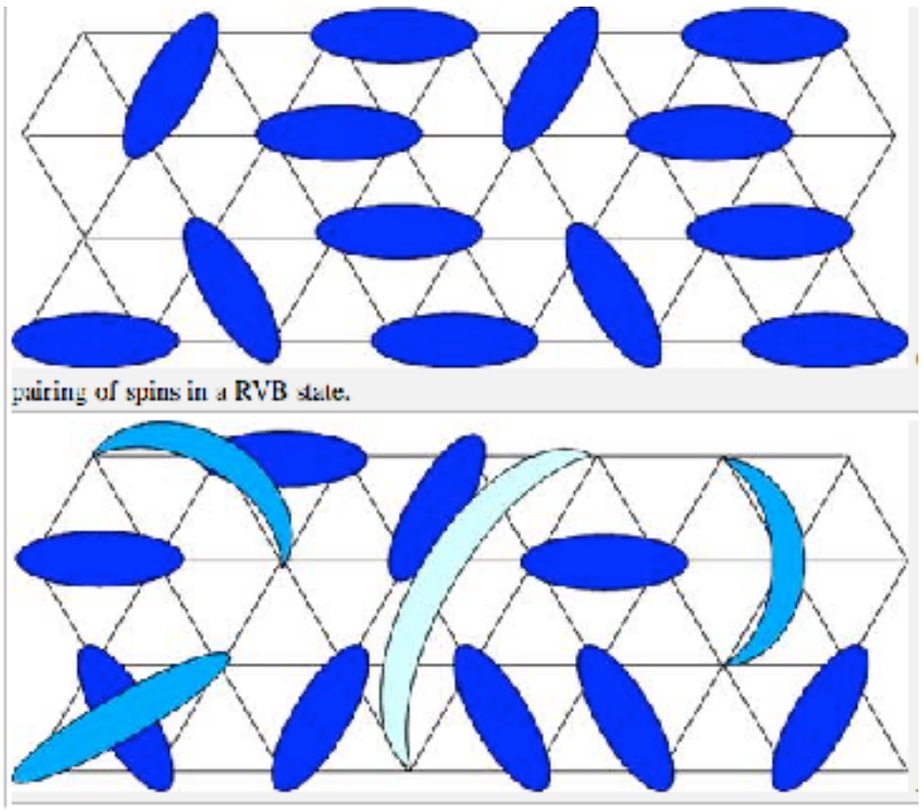
Single particle Excitation

Coldea et al PRL 2001

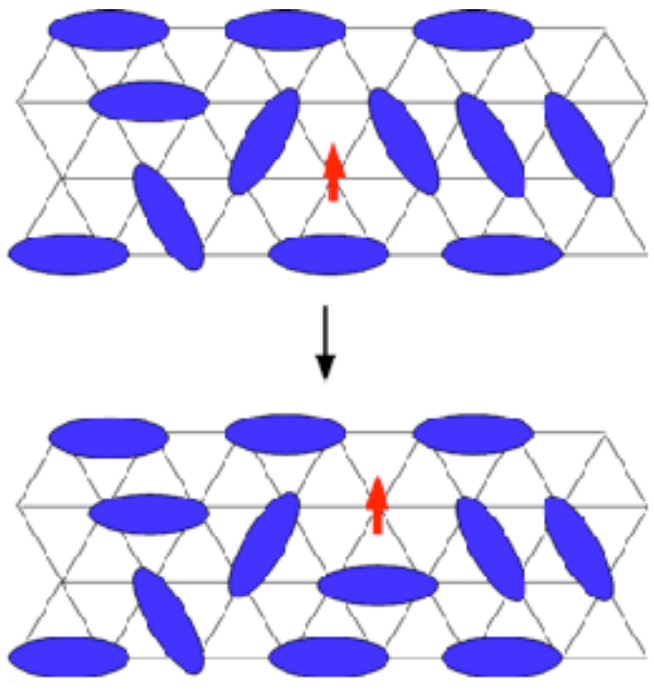
P. W. Anderson, *Resonating valence bonds: a new kind of insulator?* Mat. Res. Bul. (1973)

P. Fazekas and P. W. Anderson, Philos. Magazine (1974)

Science 235, 1196 (1987)



pairing of spins in a RVB state.

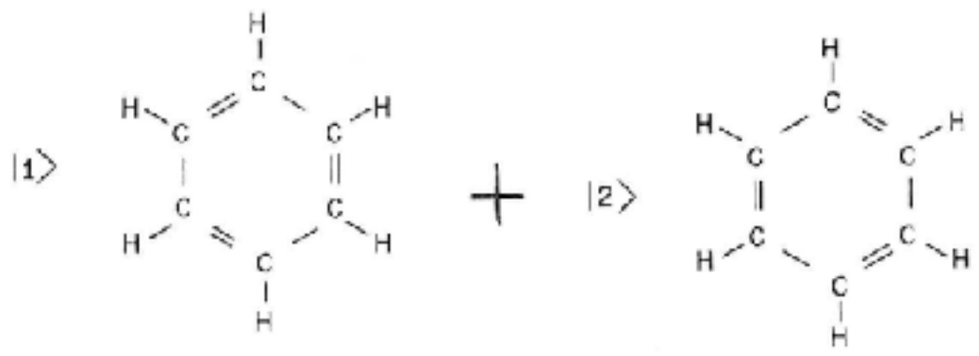


The Resonating Valence Bond State in La_2CuO_4 and Superconductivity

P. W. ANDERSON Science 235, 1196 (1987)

The oxide superconductors, particularly those recently discovered that are based on La_2CuO_4, have a set of peculiarities that suggest a common, unique mechanism: they tend in every case to occur near a metal-insulator transition into an odd-electron insulator with peculiar magnetic properties. This insulating phase is proposed to be the long-sought "resonating-valence-bond" state or "quantum spin liquid" hypothesized in 1973. This insulating magnetic phase is favored by low spin, low dimensionality, and magnetic frustration. The preexisting magnetic singlet pairs of the insulating state become charged superconducting pairs when the insulator is doped sufficiently strongly. The mechanism for superconductivity is hence predominantly electronic and magnetic, although weak phonon interactions may favor the state. Many unusual properties are predicted, especially of the insulating state.

Just like in benzene molecule (Kekule, Pauling)



excitations are obtained by breaking the singlet bond -- they are *spin-1/2 spinons*!

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Spinons are natural in d=1

Bethe's solution: 1933
identification of spinons: 1981

What is the spin of a spin wave?

L. D. Faddeev and L. A. Takhtajan

Leningrad Branch of the Steklov Mathematical Institute, Leningrad, USSR

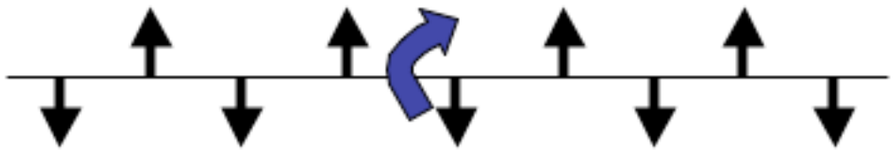
Received 15 July 1981. Available online 16 September 2002.

Abstract

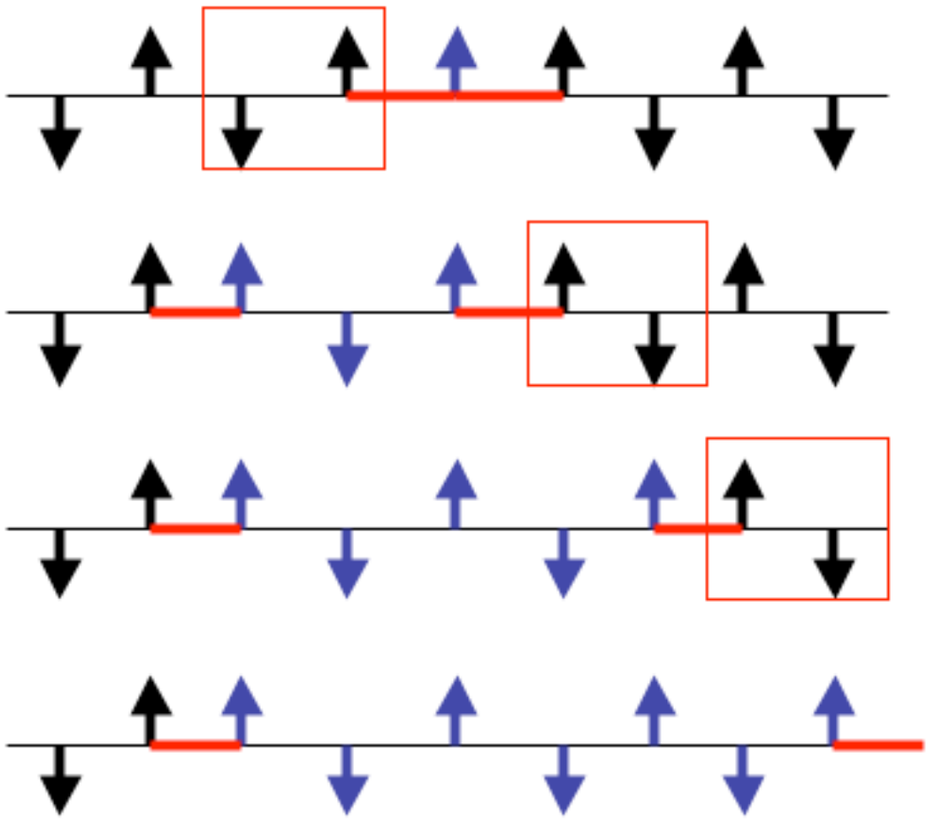
We argue that the spin of a spin wave in the Heisenberg antiferromagnetic chain of spins $\frac{1}{2}$ is equal to $\frac{1}{2}$ rather than 1 as is generally considered to be true.

Article Outline

• References



S=1 spin wave breaks into two domain walls / spinons: hence each is carrying S=1/2



Two-spinon continuum of spin-1/2 chain

Spinon energy $\omega(k_x) = \frac{\pi J}{2}|\sin(k_x)|$ de Cloizeaux-Peason dispersion, 1962

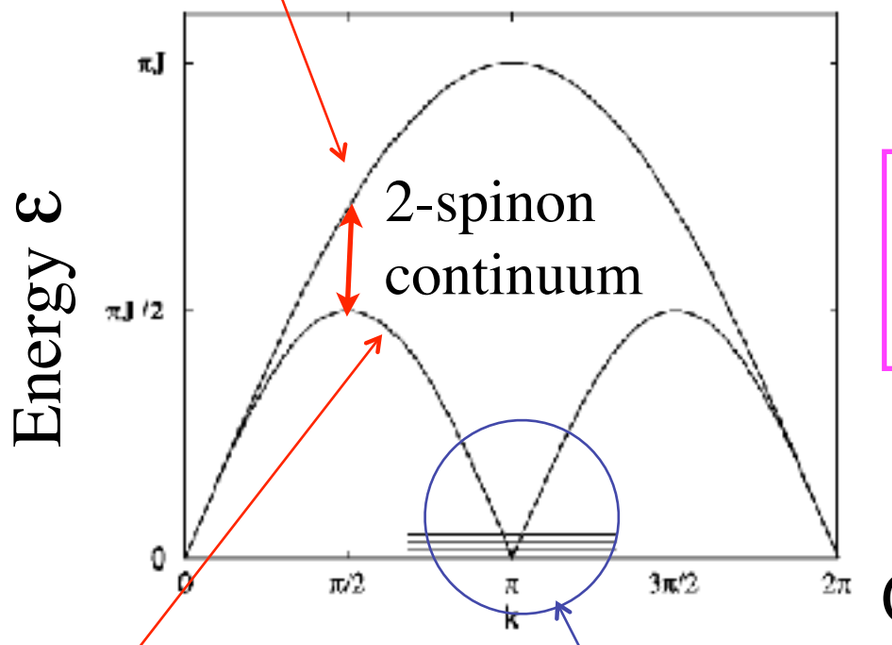
S=1 excitation $\begin{cases} \varepsilon = \omega(k_{x1}) + \omega(k_{x2}) \\ Q_x = k_{x1} + k_{x2} \end{cases}$

$$\varepsilon = \pi J \sin(\frac{Q_x}{2}) \cos(\frac{k_{x1} - k_{x2}}{2})$$

Variables:
k_{x1} and k_{x2}
or
ε and Q_x

Upper boundary

$\omega_{2,\text{upper}} = \pi J \sin(Q_x/2)$
$k_{x1} = k_{x2} = Q_x/2$

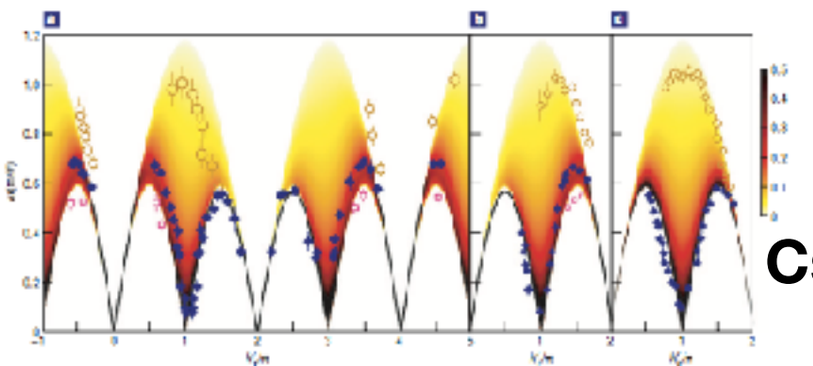


Two particle Excitation

Lower boundary

$\omega_{2,\text{lower}} = \frac{\pi J}{2} \sin(Q_x)$
$k_{x1} = 0, k_{x2} = Q_x$

Low-energy sector $Q_{AFM}=\pi$



Cs2CuCl4

ESR

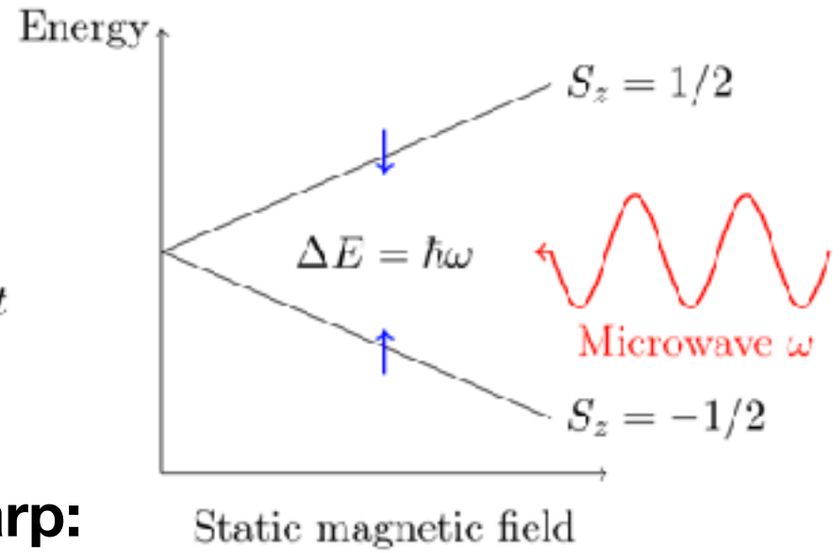
Electron Spin Resonance (ESR)

ESR measures absorption of electromagnetic radiation by a sample that is (typically) subjected to an external static magnetic field.

Linear response theory:

$$I(\vec{q}=0,\omega) = \frac{1}{2}|h|^2\omega \ \mathrm{Im}\chi_{\alpha\beta}(\vec{q}=0,\omega)$$

$$\chi_{\alpha\beta}(\vec{q}=0,\omega) = i\int_0^\infty dt \ \langle[S^\alpha(t), S^\beta(0)]\rangle e^{i\omega t}$$



For SU(2) invariant systems, completely sharp:

$$I(\omega) = \frac{1}{2}|h|^2\omega \ \delta(\omega - B)$$

No matter how exotic the ground state is!

M. Oshikawa and I. Affleck, Phys. Rev. B 65, 134410 (2002).

The key point

- Perturbations violating SU(2) symmetry do show up in ESR: line shift and line width!

- turn annoying material imperfection (spin-orbit, Dzyaloshinksii-Moriya) into a probe of exotic spin state and its excitations

- probe small-**q** excitations by ESR

absence of SU(2) is actually not a restriction!

Condensed matter physics in 21 century: the age of **spin-orbit**

✓ spintronics

✓ topological insulators, Majorana fermions

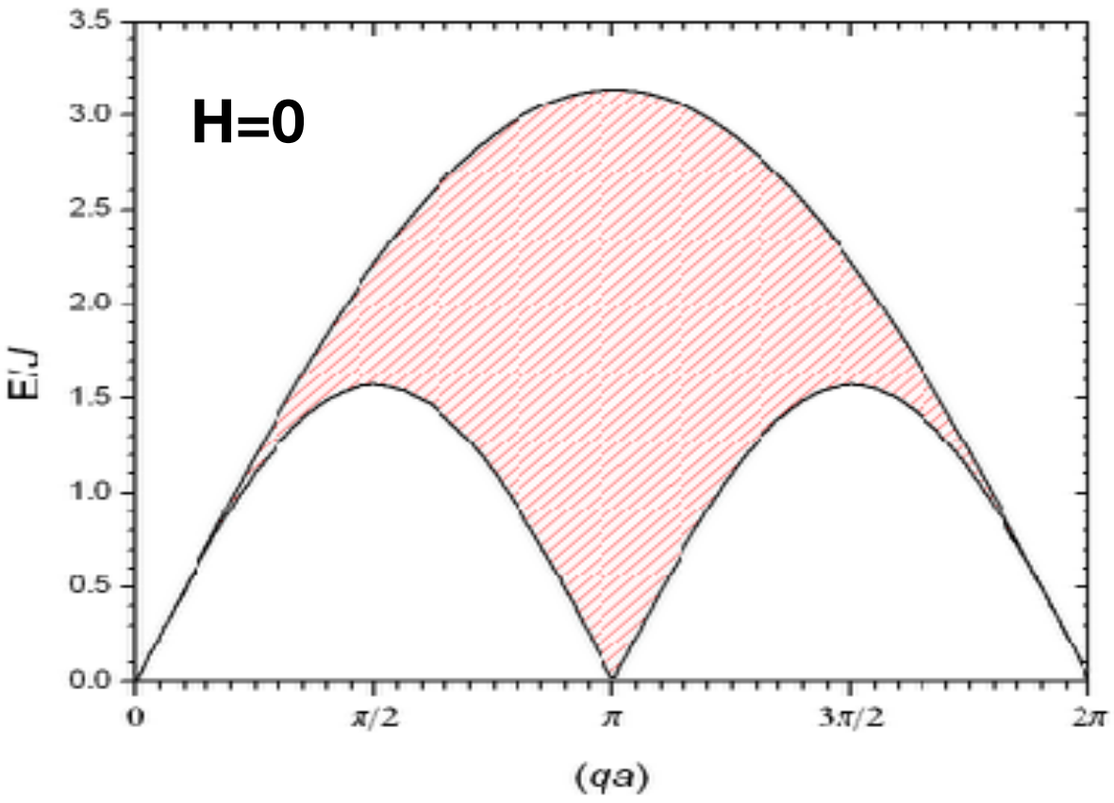
✓ Kitaev's non-abelian honeycomb spin liquid

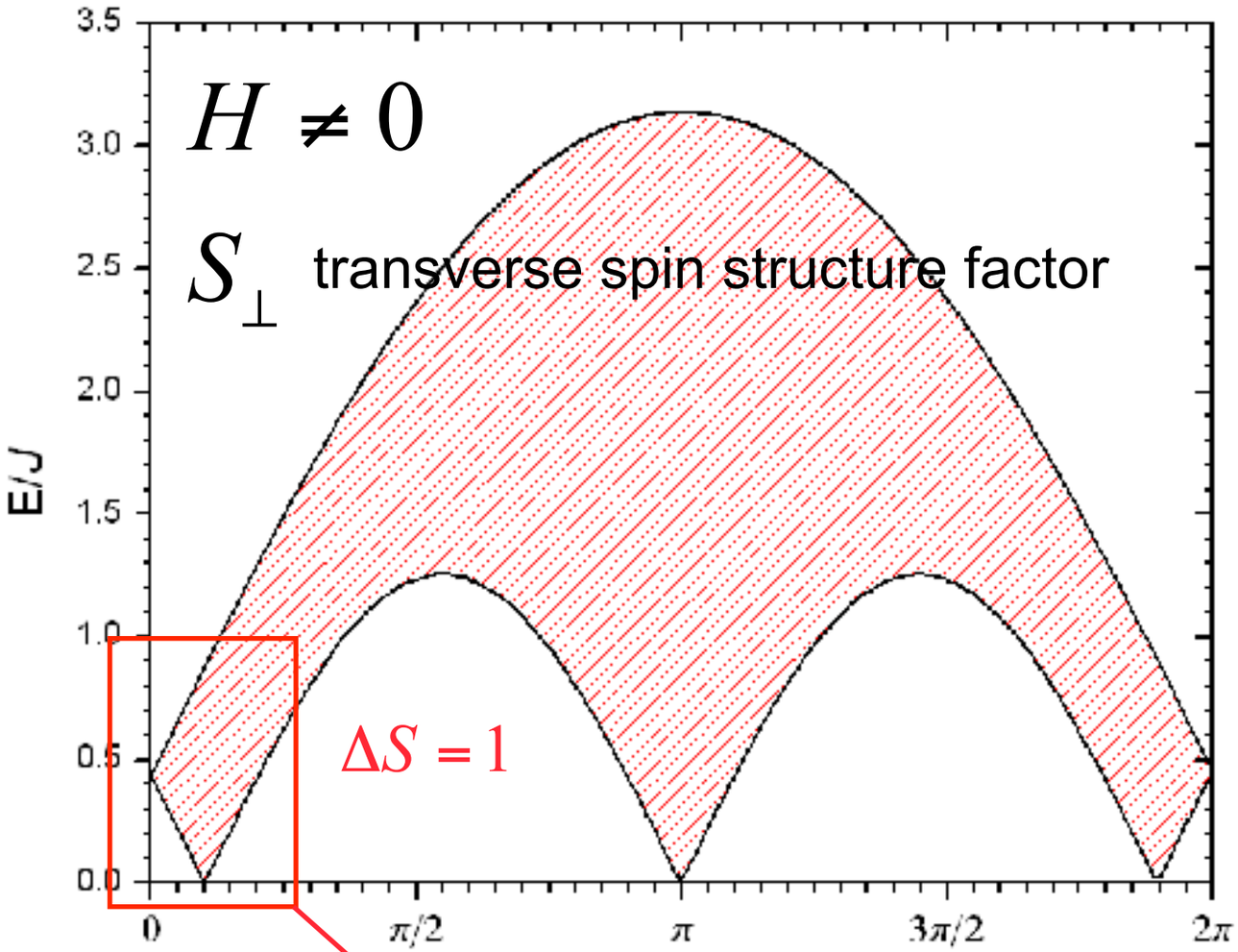
It is all about spin-orbit

Outline

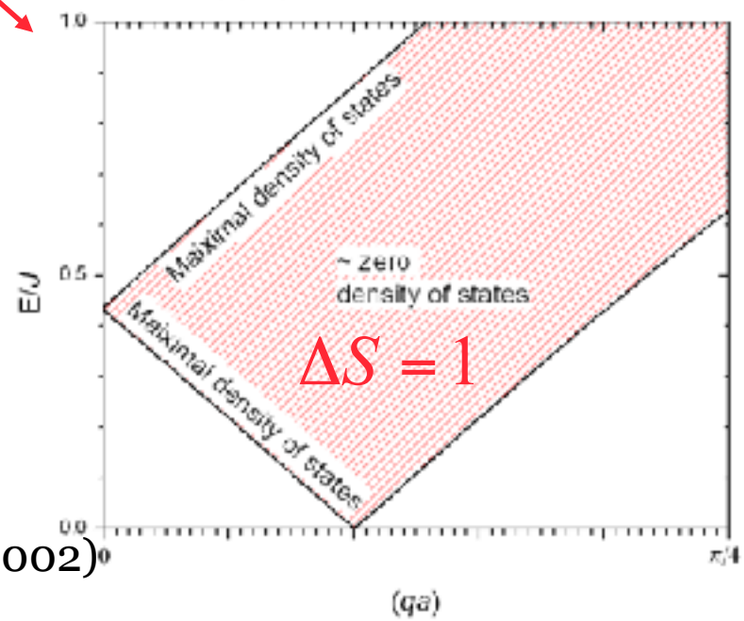
- *Main ingredients*
  - spin liquid
  - absence of spin-rotational symmetry (spin-orbit, DM, anisotropy…)
- *ESR of* Cs_2CuCl_4: one-dimensional spinon continuum
- *ESR of two-dimensional spinon continuum YbMgGaO4*
- Conclusions

Probing spinon continuum in one dimension





$$I(\omega) \propto \delta(\omega - H)$$



Dender et al, PRL 1997

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

Uniform Dzyaloshinskii-Moriya interaction

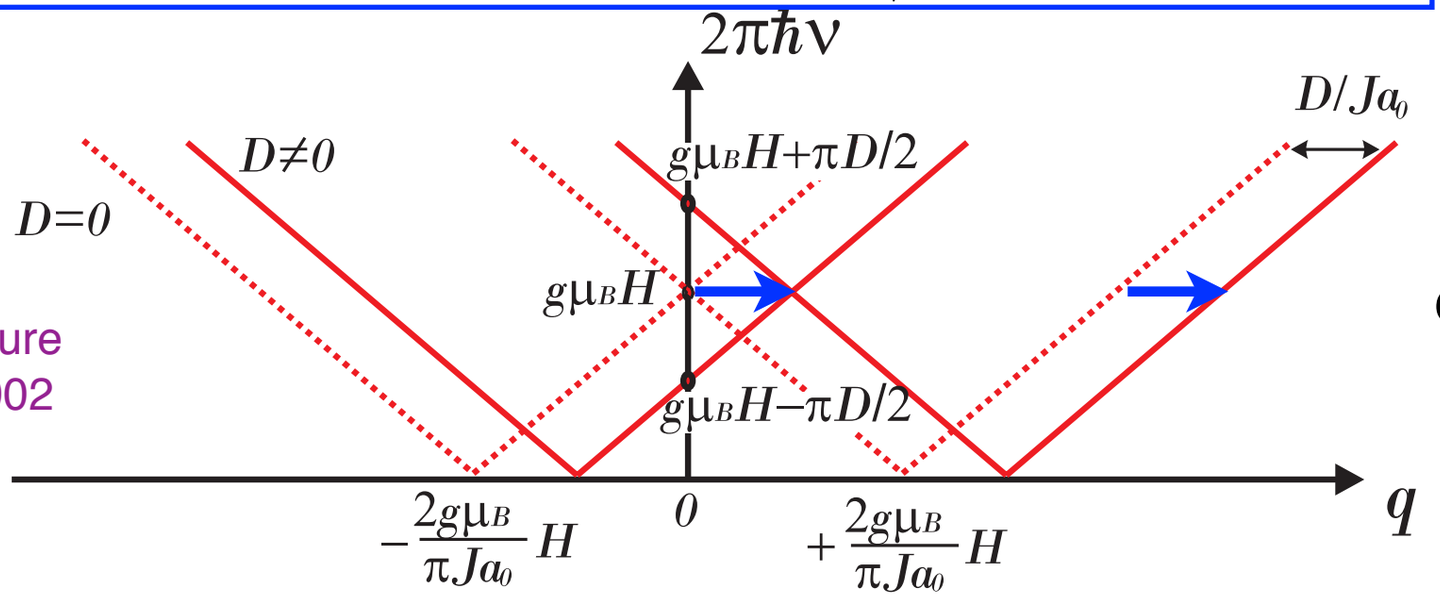
$$H = \sum_{x,y,z} \boxed{JS_{x,y,z} \cdot S_{x+1,y,z}} - \boxed{D_{y,z} \cdot S_{x,y,z} \times S_{x+1,y,z}} - g\mu_B H \cdot S_{x,y,z}$$

chain — uniform DM along the chain — magnetic field

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

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

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



dotted lines: D=0 picture
Oshikawa, Affleck 2002

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

H ∥ D

DM interaction allows to probe spinon continuum at finite "boost" momentum

Cs_2CuCl_4 ESR data: T=1.3 K

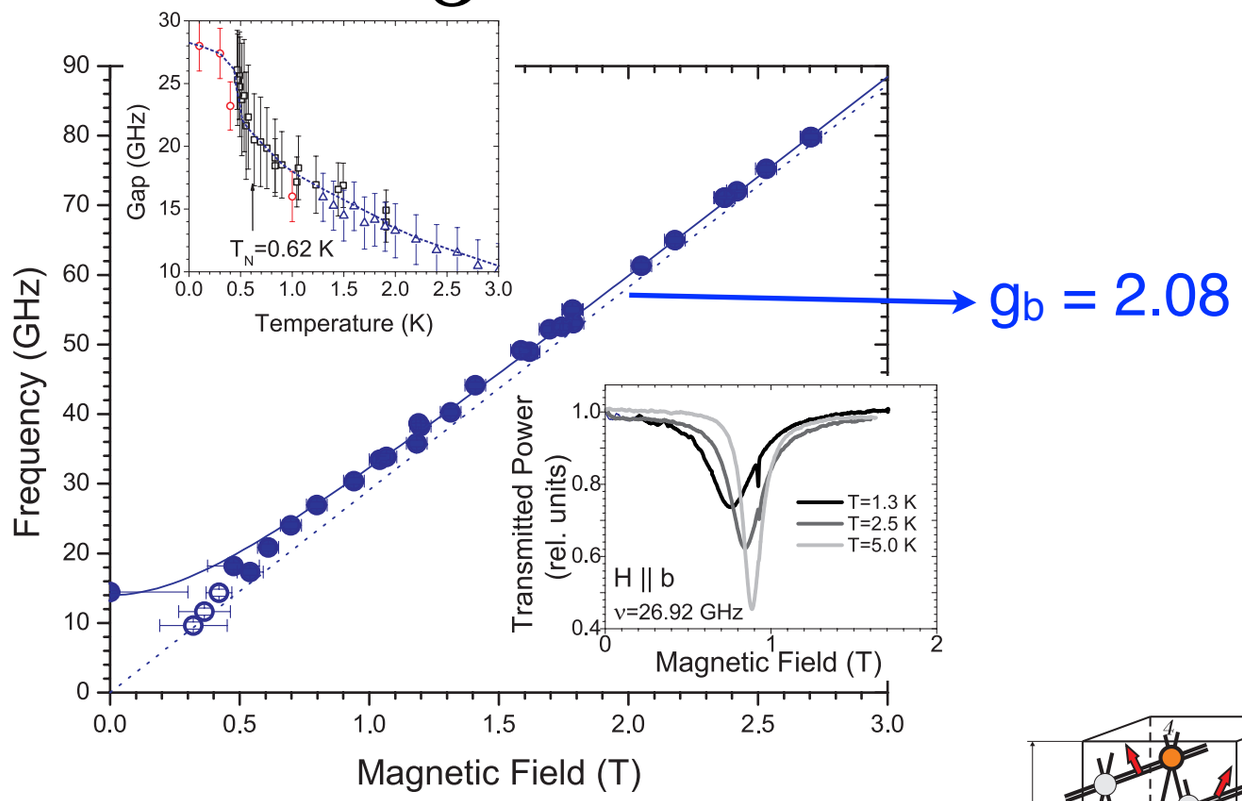
Povarov et al, 2011

- H along b-axis

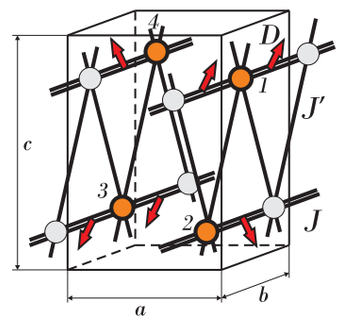
✓ **gap**-like behavior for ν > 17 GHz

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

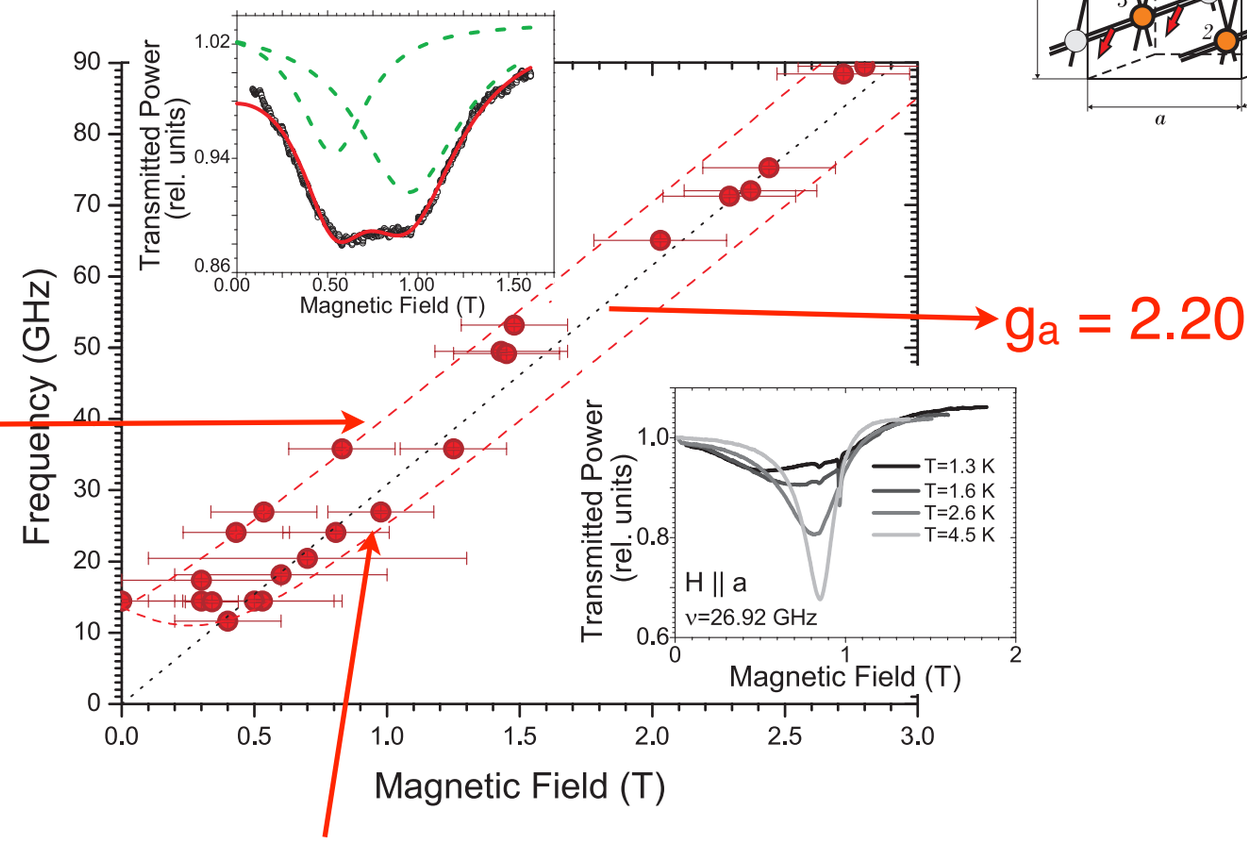
✓ loss of intensity for ν < 17 GHz



$g_b = 2.08$



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



$g_a = 2.20$

Cs2CuCl4 ESR data

PRL 107, 037204 (2011) PHYSICAL REVIEW LETTERS week ending 15 JULY 2011

Modes of Magnetic Resonance in the Spin-Liquid Phase of Cs_2CuCl_4

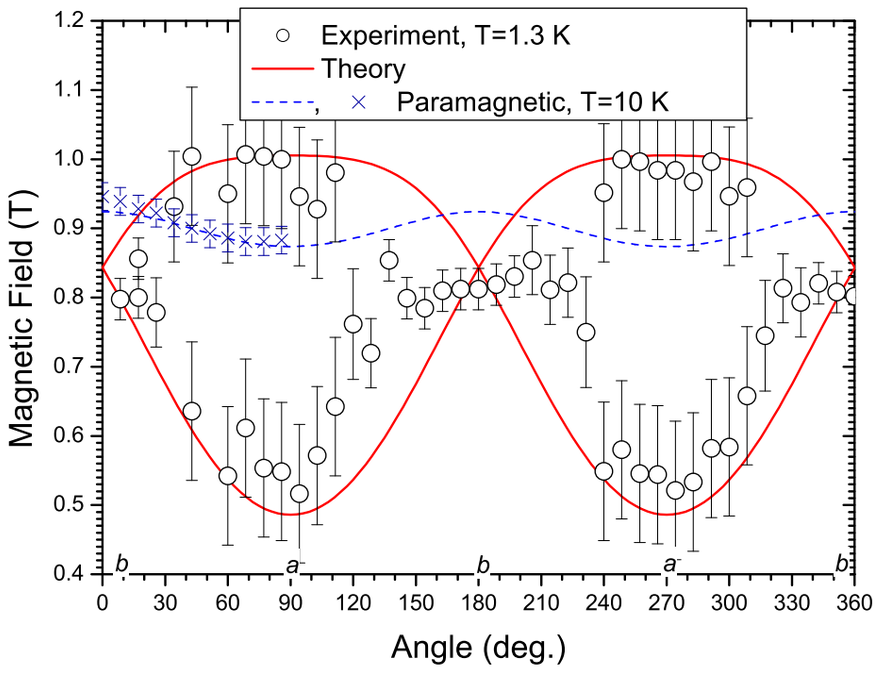
K. Yu. Povarov,[1,*] A. I. Smirnov,[1,2] O. A. Starykh,[3,4] S. V. Petrov,[1] and A. Ya. Shapiro[5]

- 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

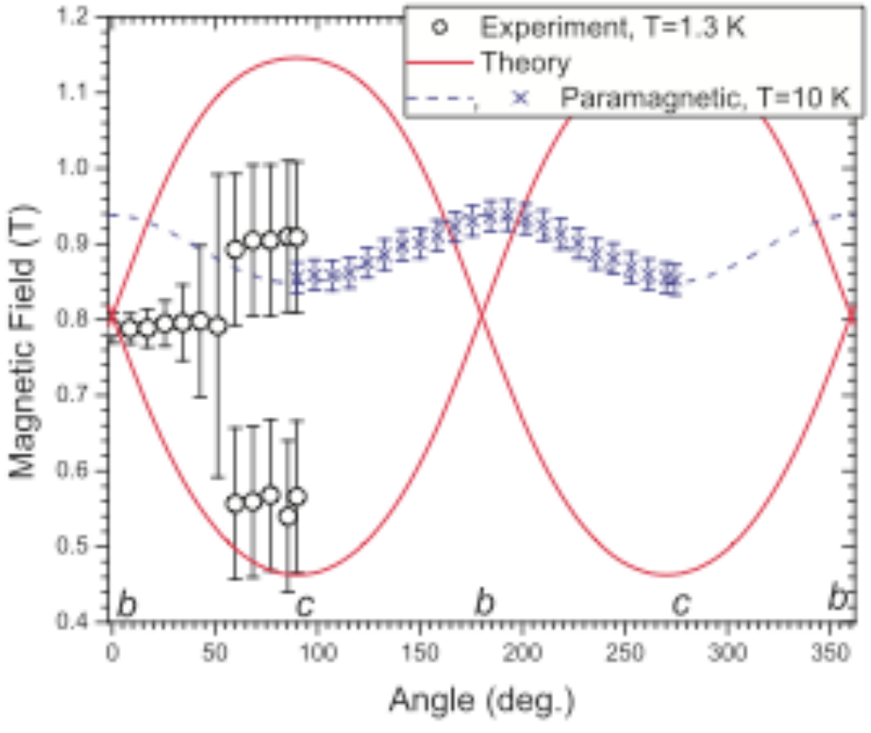
D ~ J/10

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

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

T-linear line width
S. C. Furuya Phys. Rev. B 95, 014416 (2017)

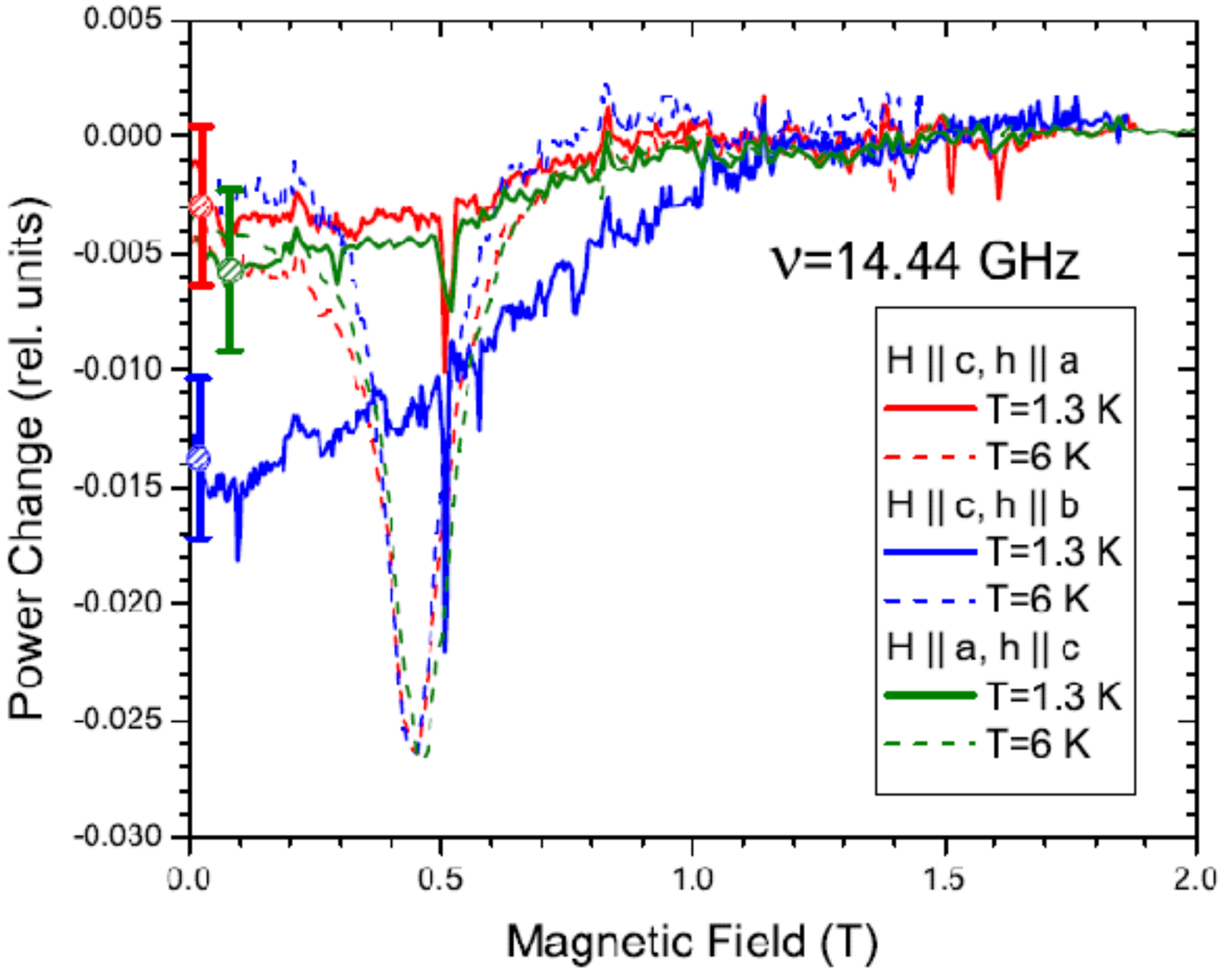
b-c plane

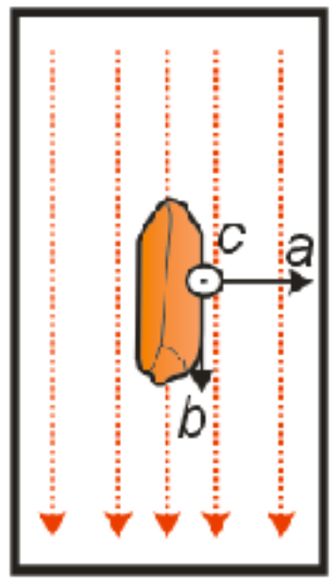


Zero-field absorption:

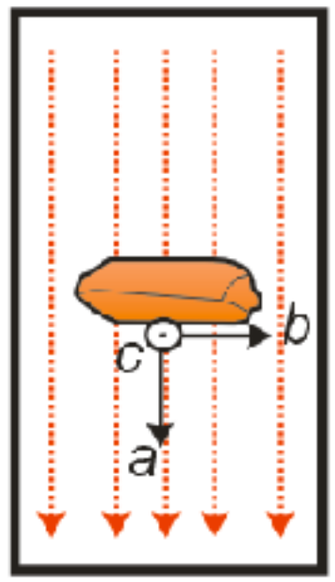
✓ 1) ESR absorption in the absence of H $\nu \sim \sqrt{D_a^2 + D_c^2}$

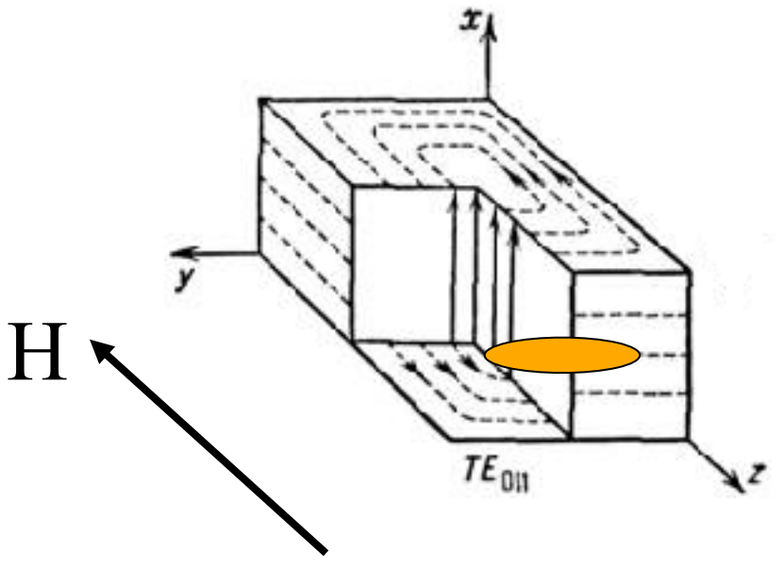
✓ 2) strong polarization dependence in zero field





h





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] Povarov et al, PRL 2011

PHYSICAL REVIEW B 92, 134417 (2015)

Electron spin resonance in a model $S = \frac{1}{2}$ chain antiferromagnet with a uniform Dzyaloshinskii-Moriya interaction

A. I. Smirnov,[1] T. A. Soldatov,[1,2] K. Yu. Povarov,[3,*] M. Hälg,[3] W. E. A. Lorenz,[3] and A. Zheludev[3]
[1]*P. L. Kapitza Institute for Physical Problems, RAS, 119334 Moscow, Russia*
[2]*Moscow Institute for Physics and Technology, 141700 Dolgoprudny, Russia*
[3]*Neutron Scattering and Magnetism, Laboratory for Solid State Physics, ETH Zürich, Switzerland*
(Received 27 July 2015; revised manuscript received 6 October 2015; published 22 October 2015)

The electron spin resonance spectrum of a quasi-1D $S = 1/2$ antiferromagnet $K_2CuSO_2Br_2$ was found to demonstrate an energy gap and a doublet of resonance lines in a wide temperature range between the Curie-Weiss and Neël temperatures. This type of magnetic resonance absorption corresponds well to the two-spinon continuum of excitations in $S = 1/2$ antiferromagnetic spin chain with a uniform Dzyaloshinskii-Moriya interaction between the magnetic ions. A resonance mode of paramagnetic defects demonstrating strongly anisotropic behavior due to interaction with spinon excitations in the main matrix is also observed.

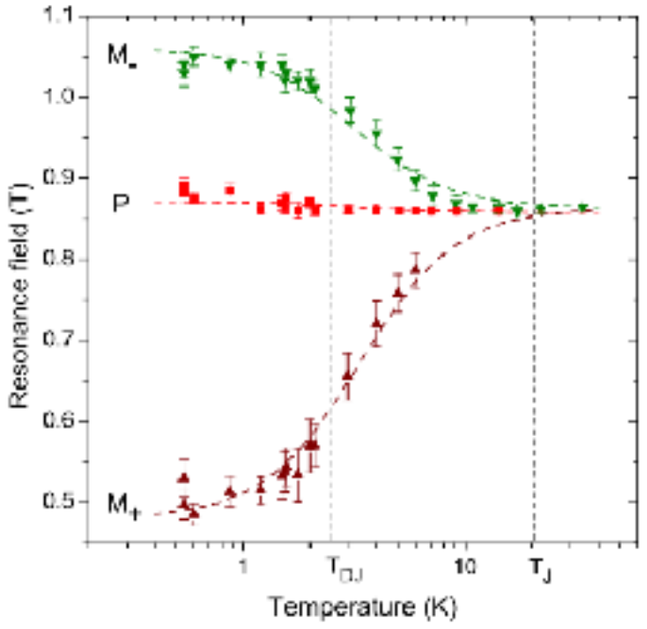


FIG. 4. (Color online) Temperature dependence of 27.83 GHz ESR fields at $\mathbf{H} \parallel b$ for the components of the doublet M_-, M_- and of the paramagnetic line P, associated with the defects. Characteristic temperatures marked on the horizontal axis are $T_{DJ} = \sqrt{DJ}/k_B$ and $T_J = J/k_B$. Dashed lines are guide to the eyes.

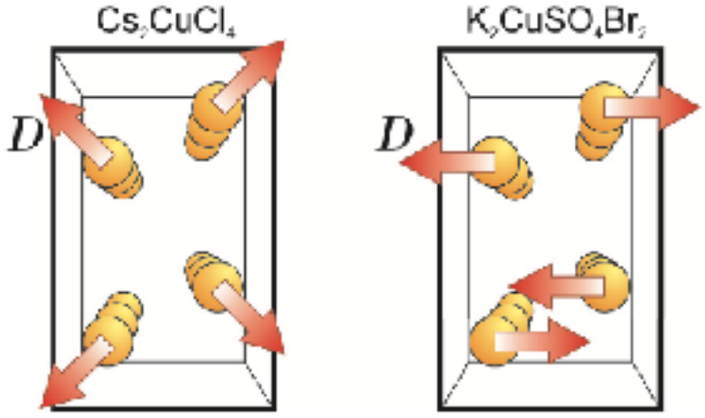


FIG. 2. (Color online) Comparison of the Dzyaloshinskii-Moriya vectors in Cs_2CuCl_4 and $K_2CuSO_2Br_2$. Perspective view along the spin chains.

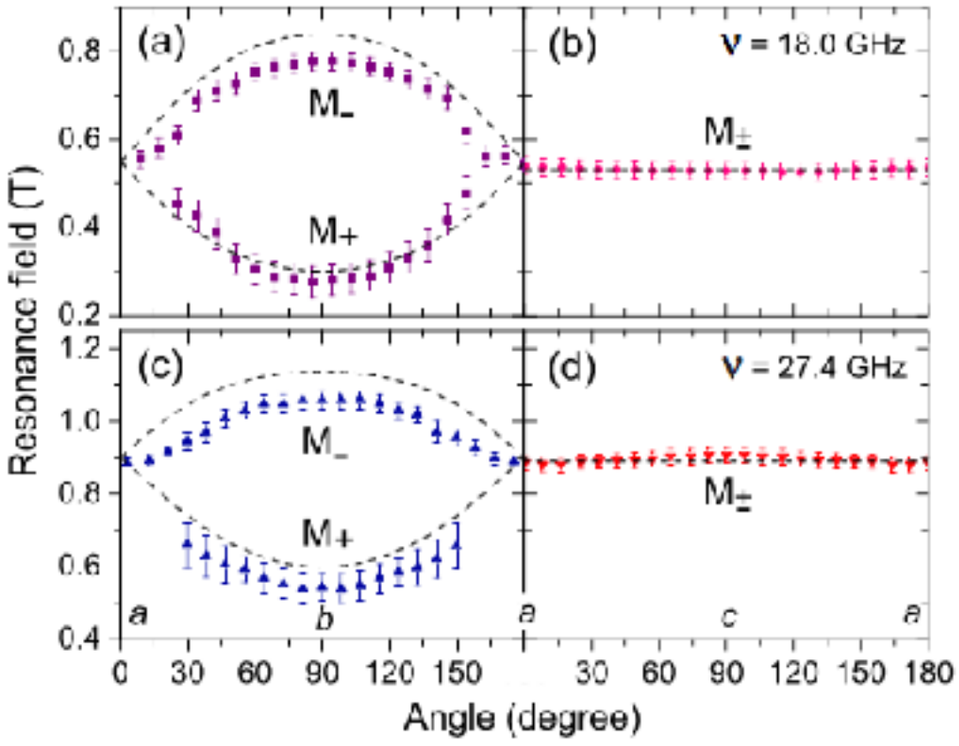
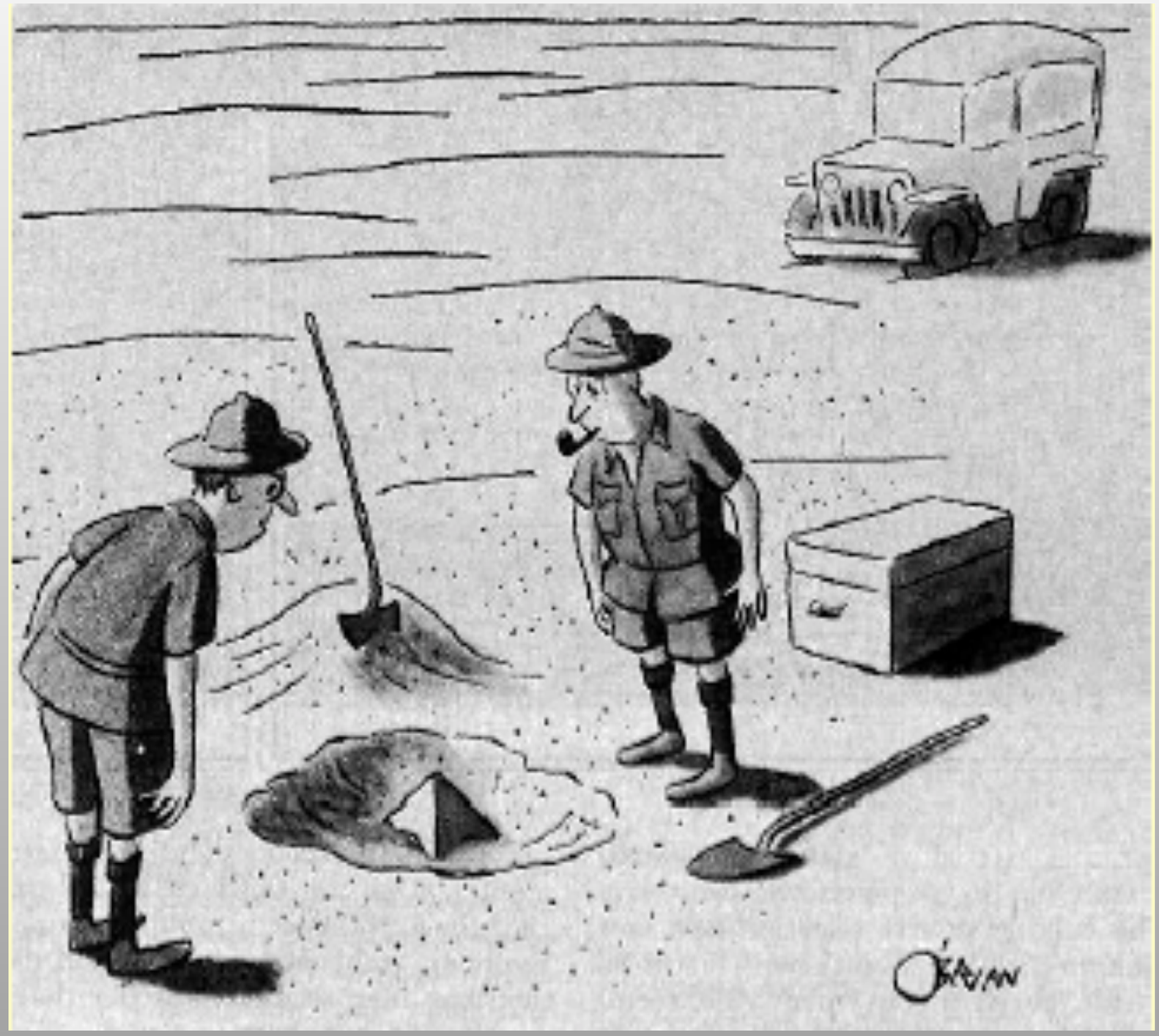


FIG. 8. (Color online) Angular dependencies of the resonant fields at two frequencies for $T = 1.3$ K. 18.0 GHz ESR fields for field in ab plane (upper left panel), and in ac plane (upper right panel). 27.4 GHz ESR fields for field in ab plane (lower left panel), and in ac plane (lower right panel). Dashed lines are theoretical dependencies according to (2).

J = 20 K, D = 0.27 K

Two-dimensional spin liquids with fractionalized excitations



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

Two (and three) dimensional spin liquids

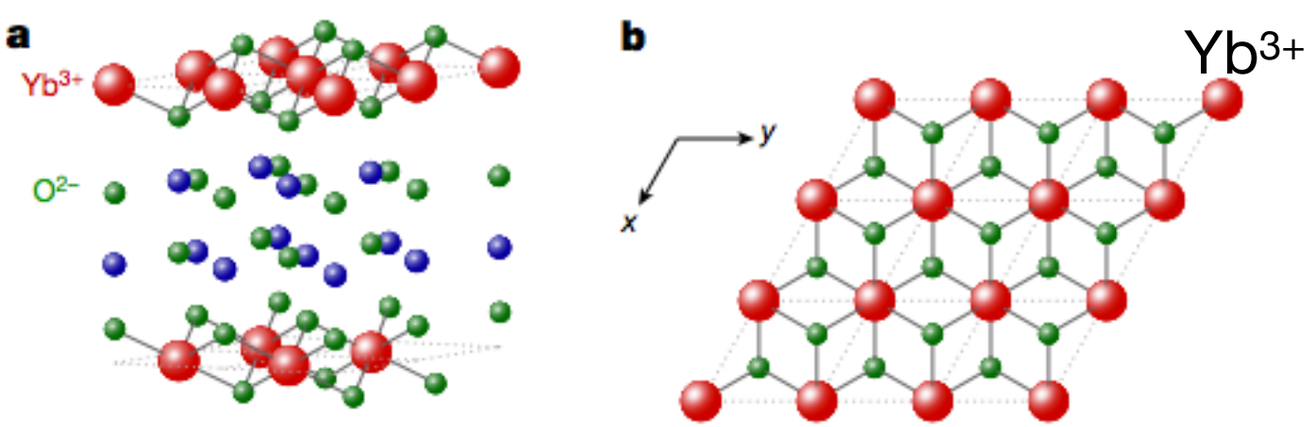
No magnetic order
Significant spin-orbit interaction
Fractionalized excitations

↓

Kitaev materials
Pyrochlores
Iridates

↓

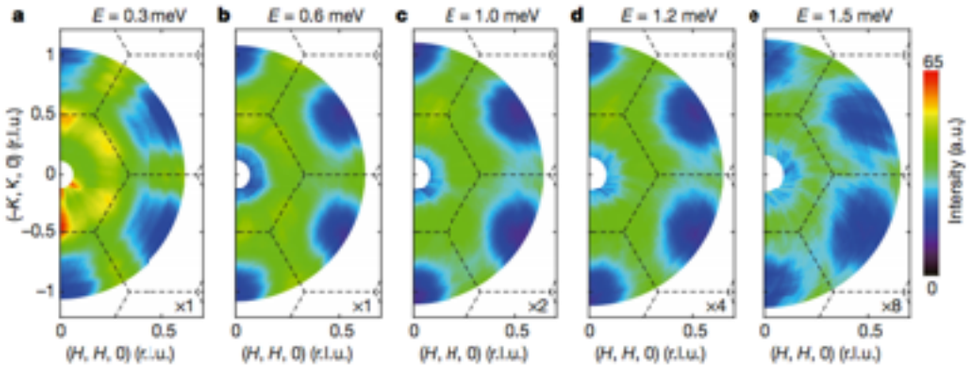
YbMgGaO4
interesting
promising
simple lattice

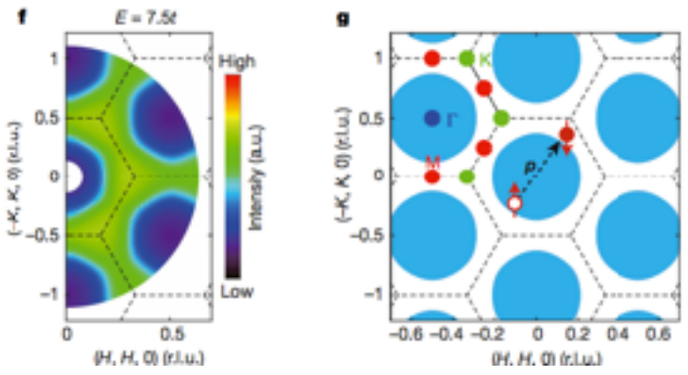


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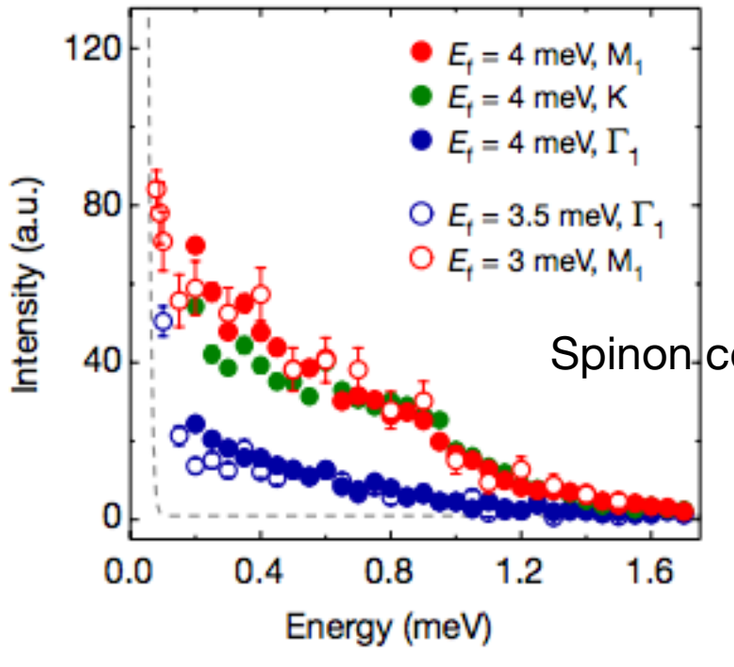
Evidence for a spinon Fermi surface in a triangular-lattice quantum-spin-liquid candidate

Yao Shen[1], Yao-Dong Li[2], Hongliang Wo[1], Yuesheng Li[1], Shoudong Shen[1], Bingying Pan[1], Qisi Wang[1], H. C. Walker[4], P. Steffens[5], M. Boehm[5], Yiqing Hao[1], D. L. Quintero-Castro[6], L. W. Harriger[7], M. D. Frontzek[8], Lijie Hao[9], Siqin Meng[9], Qingming Zhang[9,10,11], Gang Chen[1,11,12] & Jun Zhao[1,11]





Sign of spinon Fermi surface?

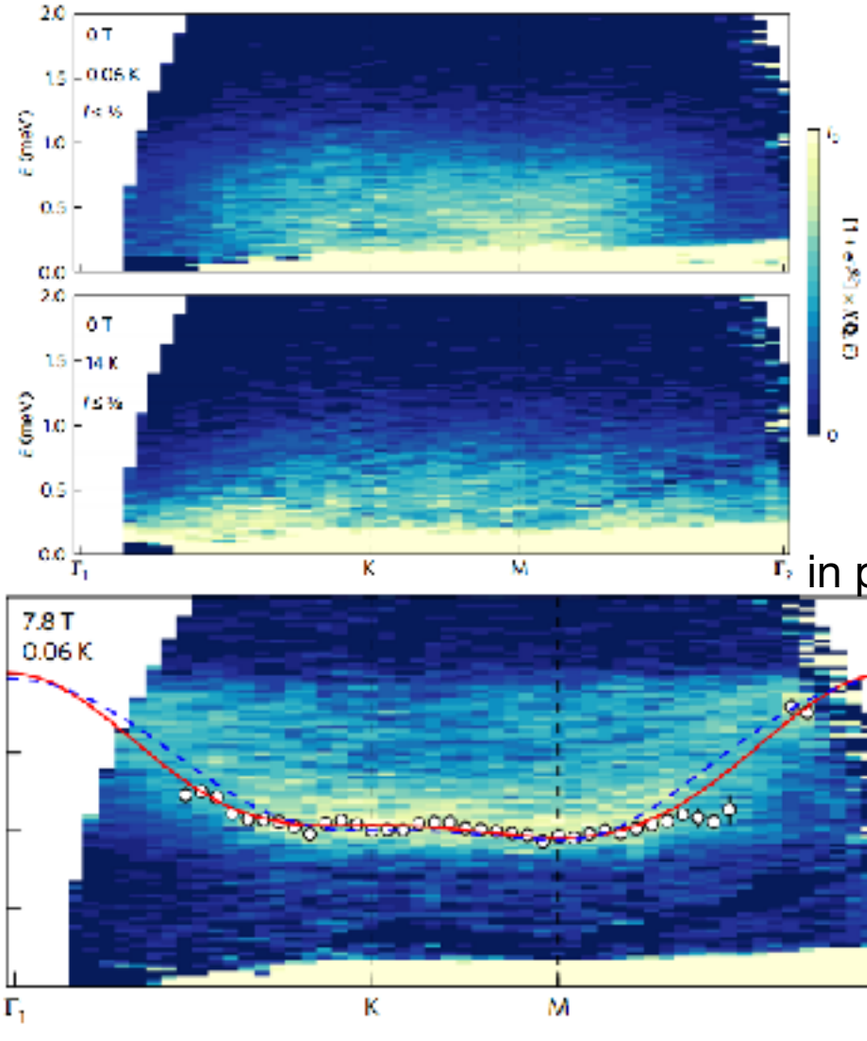


Spinon continuum?

nature physics
LETTERS
PUBLISHED ONLINE: 5 DECEMBER 2016 | DOI: 10.1038/NPHYS3971

Continuous excitations of the triangular-lattice quantum spin liquid $YbMgGaO_4$

Joseph A. M. Paddison[1], Marcus Daum[1†], Zhiling Dun[2†], Georg Ehlers[3], Yaohua Liu[3], Matthew B. Stone[3], Haidong Zhou[2] and Martin Mourigal[1*]



Broad signal in polarized phase?

very disordered

$$\mathcal{H} = \sum_{\langle ij \rangle} \left[J_1^{zz} S_i^z S_j^z + J_1^{\pm} \left(S_i^+ S_j^- + S_i^- S_j^+ \right) + J_1^{\pm\pm} \left(\gamma_{ij} S_i^+ S_j^+ + \gamma_{ij}^* S_i^- S_j^- \right) \right]$$
$$+ \sum_{\langle\langle ij \rangle\rangle} \left[J_2^{zz} S_i^z S_j^z + J_2^{\pm} \left(S_i^+ S_j^- + S_i^- S_j^+ \right) \right]$$
$$- \mu_0 \mu_B \sum_i \left[g_{\perp} \left(H^x S_i^x + H^y S_i^y \right) + g_{\parallel} H^z S_i^z \right] \qquad (1)$$

Spinon hypothesis

arxiv:1612.03447

The Spinon Fermi Surface U(1) Spin Liquid in a Spin-Orbit-Coupled Triangular Lattice Mott Insulator $YbMgGaO_4$

Yao-Dong Li[1], Yuan-Ming Lu[2], and Gang Chen[1,3]*

PHYSICAL REVIEW LETTERS 120, 037204 (2018)

Spinon Magnetic Resonance of Quantum Spin Liquids

Zhu-Xi Luo,[1,*] Ethan Lake,[1] Jia-Wei Mei,[2] and Oleg A. Starykh[1,†]

Spinon mean-field Hamiltonian
derived with the help of
Projective Symmetry Group (PSG) analysis

$$S^a_{\mathbf{r}} = \frac{1}{2} f^\dagger_{\mathbf{r}\alpha} \sigma^a_{\alpha\beta} f_{\mathbf{r}\beta}$$

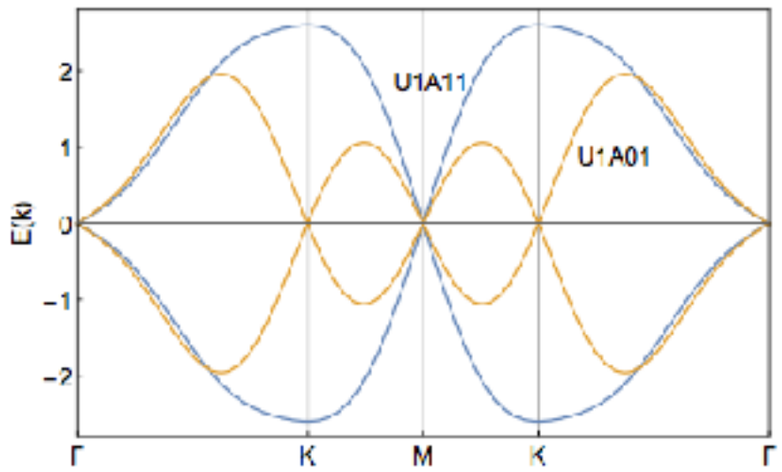


FIG. S-5. Spinon dispersions $E_{1,2}(\mathbf{k})$ along the line Γ-K-M-K-Γ for U1A11 and U1A01 states.

Dirac spectrum!

Basic idea: physical spin S is bilinear of spinons f,
spinons have bigger symmetry group than spins,
this leads to gauge freedom and
different classes of possible mean-fields.
These classes describe the same spin problem.

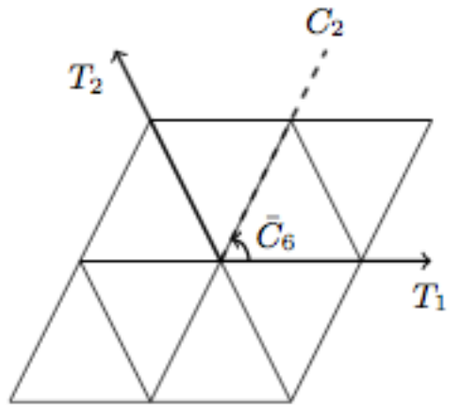
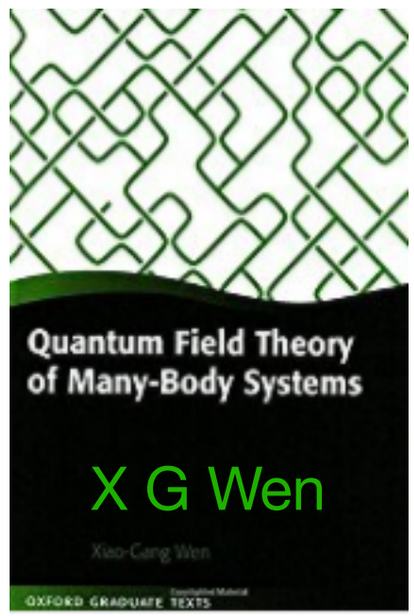
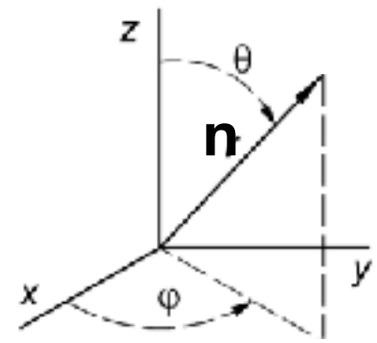


FIG. S-1. The symmetry operations.



Spinon magnetic resonance

AC magnetic field couples to the total spin $S^a_{\mathbf{r}} = \frac{1}{2} f^\dagger_{\mathbf{r}\alpha} \sigma^a_{\alpha\beta} f_{\mathbf{r}\beta}$

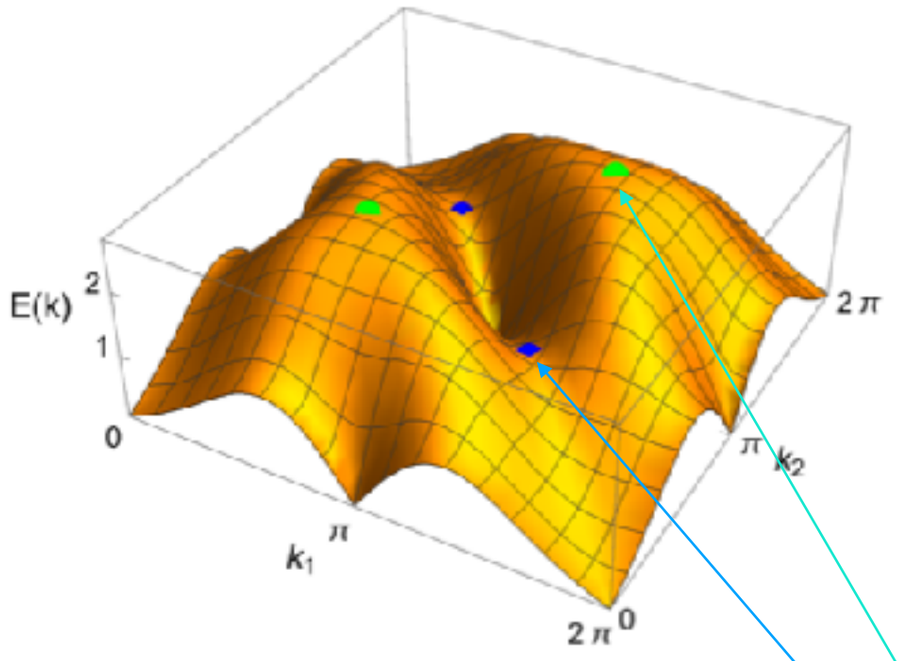


$$V(t) = he^{-i\omega t}\mathbf{n} \cdot \frac{1}{2} \sum_{\mathbf{r}} (f^\dagger_{\mathbf{r}\uparrow}, f^\dagger_{\mathbf{r}\downarrow}) \boldsymbol{\sigma} \begin{pmatrix} f_{\mathbf{r}\uparrow} \\ f_{\mathbf{r}\downarrow} \end{pmatrix}$$

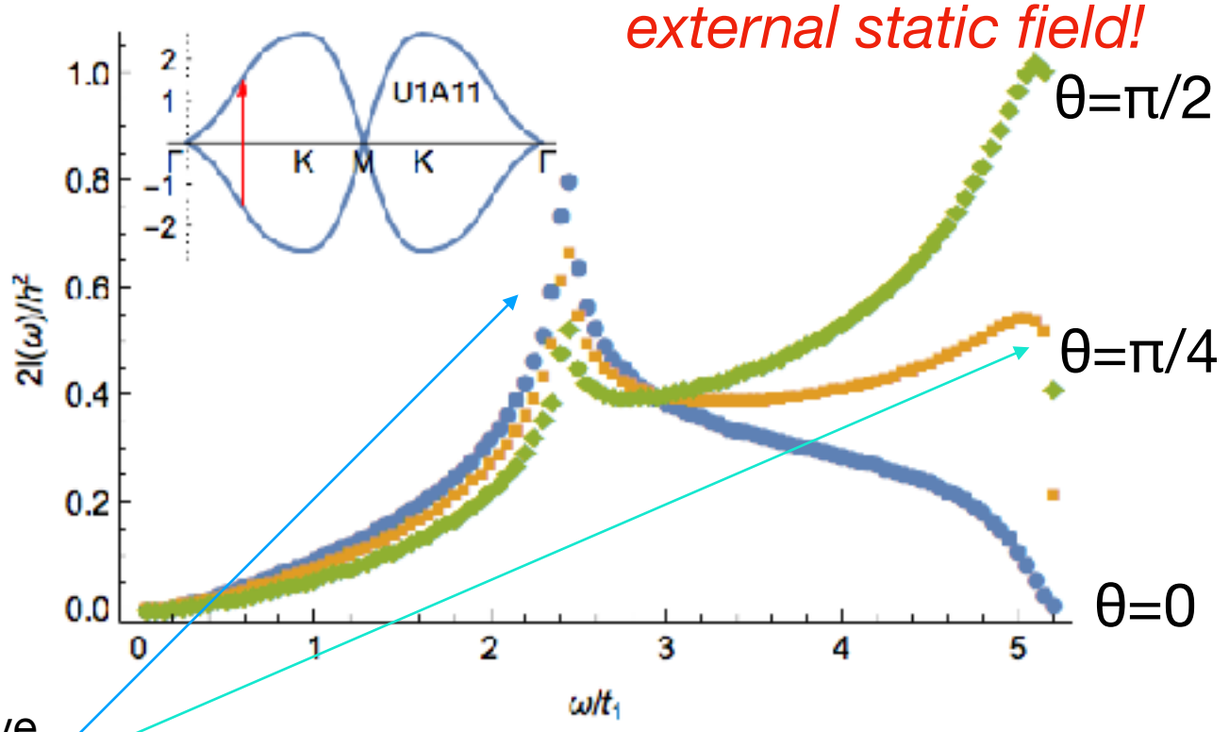
Rate of energy absorption $I(\omega) = -\omega \chi''_{\mathrm{nn}}(\omega) |h|^2/2$

Dynamic susceptibility at **q**=0

$$\chi_{\mathrm{nn}}(\omega) = \frac{1}{4N} \sum_{\mathbf{k}} \frac{n_{\mathbf{k}\alpha} - n_{\mathbf{k}\beta}}{\omega + E_\alpha(\mathbf{k}) - E_\beta(\mathbf{k}) + i0} \times (U^+_{\mathbf{k}} \sigma^a U_{\mathbf{k}})_{\alpha\beta} (U^+_{\mathbf{k}} \sigma^b U_{\mathbf{k}})_{\beta\alpha} \hat{n}^a \hat{n}^b$$

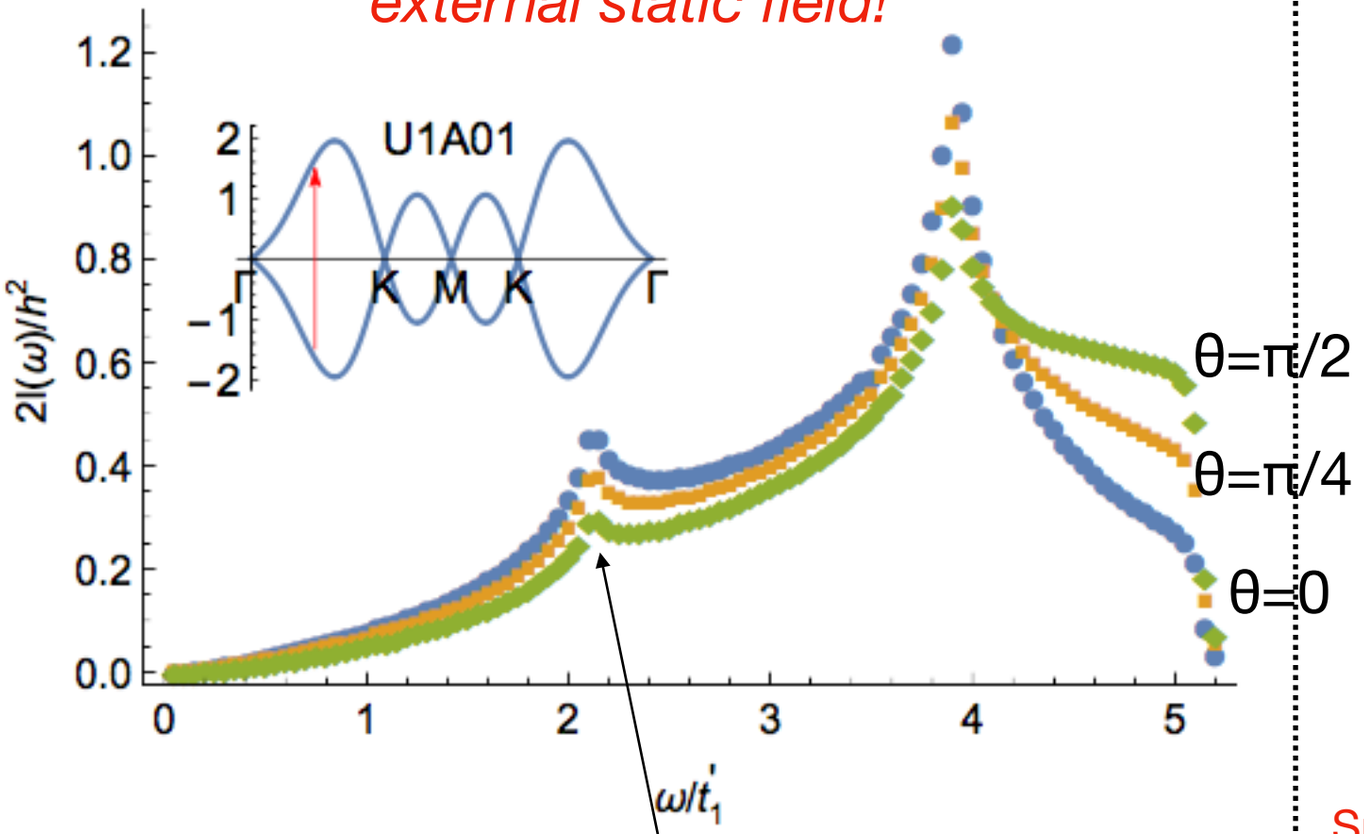


Absorption without external static field!

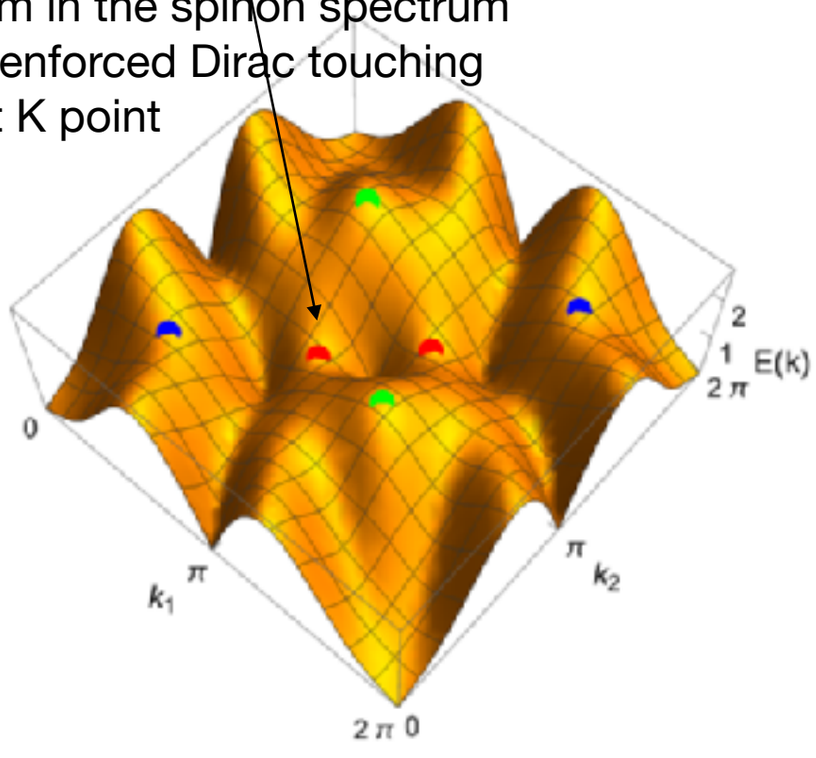


van Hove singularities

Absorption without external static field!

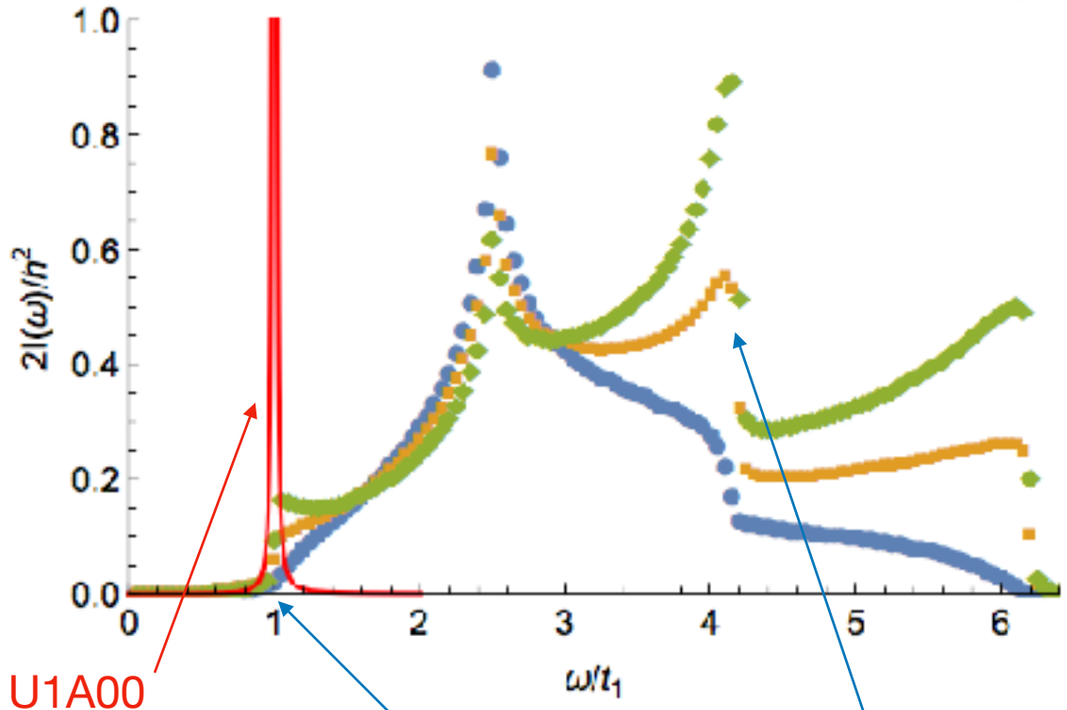


Additional extremum in the spinon spectrum due to symmetry-enforced Dirac touching at K point



With magnetic field along Z

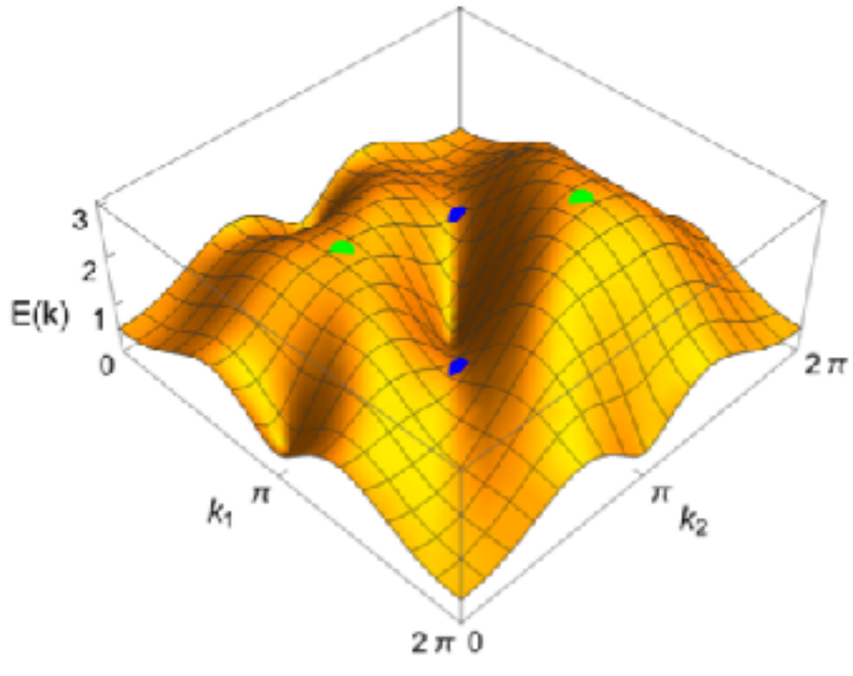
U1A11



U1A00
Spinon Fermi surface state
$\sin^2\theta\ \delta(\omega - B_z)$

threshold frequency is determined by B_z

additional singularity



Existing ESR in YbMgGaO4

Y. Li, G. Chen, W. Tong et al,
Phys. Rev. Lett. 115, 167203 (2015).

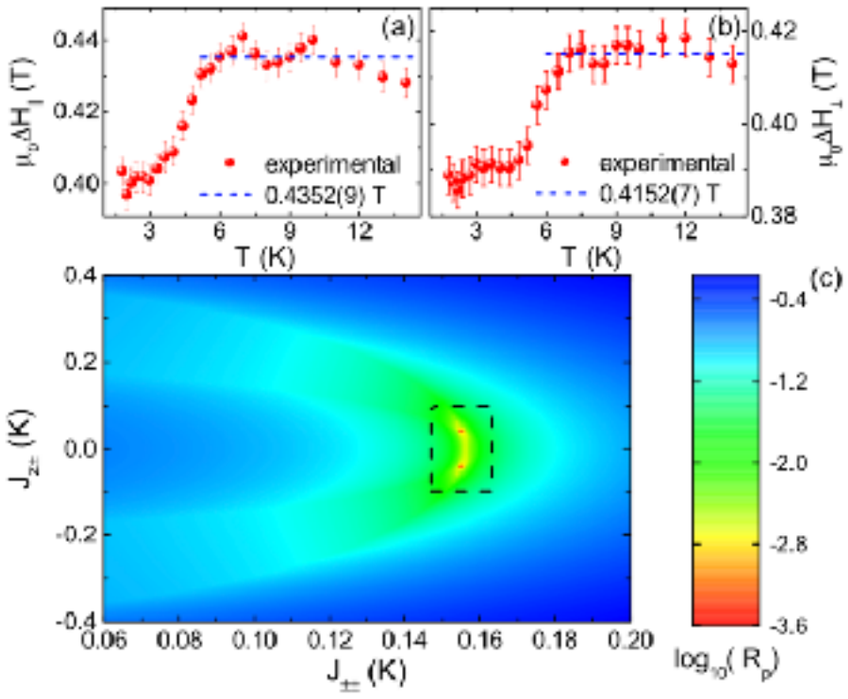


FIG. 3. (Color online.) The temperature dependence of ESR linewidths (a) parallel and (b) perpendicular to the c axis. The dashed lines are the corresponding constant fits to the ESR linewidth data at $T > 6$K. (c) The deviation, R_p, of the experimental ESR linewidthes from the theoretical ones for $YbMgGaO_4$. The dashed rectangle gives the optimal parameters $|J_{\pm\pm}| = 0.155(9)$K and $|J_{z\pm}| = 0.04(10)$K.

Minimum temperature: 1.8 K

X. Zhang, F. Mahmood, M. Daum et al,
arXiv: 1708.07503.

Model	A	B	B*	C
$J_1^{\pm\pm}$ (meV)	0.126	0.164	0.151(5)	0.149(5)
$J_1^{\pm}$ (meV)	0.109	0.108	0.088(3)	0.085(3)
$J_1^{z\pm}$ (meV)	0.013	0.056	0.13(2)	0.07(6)
$\lvert J_1^{z\pm}\rvert$ (meV)	0	0.098	0.1(1)	0.1(1)
J_2/J_1	0.22	0	0	0.18(7)
$g_{\parallel}$	3.72	3.72	3.81(4)	3.81(4)
$g_{\perp}$	3.06	3.06	3.53(5)	3.53(5)

TABLE I. Exchange parameters for different models derived from fitting the spin-wave dispersions. Models A and B are from [60] and [43], respectively. Model C is from our global fit to the TDTS and INS data. Model B* is from a global fit to the data by ignoring NNN interactions, i.e. $J_2 = 0$. Uncertainties in the values represent the 99.7% confidence interval (3 s.d.) in extracting the fitting parameters.

Lower the temperature to see the spinon effect!
T ~ 0.1 K

Conclusion:

Spinon magnetic resonance is generic feature of spin liquids with significant spin-orbit interaction and fractionalized excitations

Main features:

- **broad continuum response**
- **zero-field absorption**
- **strong polarization dependence**
- **van Hove singularities of spinon spectrum**



Work in progress:
Two-dimensional spinon Fermi surface with DM interaction -
ESR line width dependence on T, h is due to gauge fluctuations.

OS, Leon Balents

BigMag@UCSB

A workshop to identify transformational science that would be enabled by coupling a ~30 T magnet to UCSB's Terahertz Free-Electron Lasers

ESR response of many interesting materials is in Terahertz range

Absorption is present even *without* the field

High magnetic field allows to track quantum fluctuations as they develop with *lowering* of the magnetic field

Explore temperature/magnetic field/polarization dependences.