Breaking the spin waves: spinons in Cs_2CuCl_4 and elsewhere

Oleg Starykh, University of Utah

In collaboration with:

K. Povarov, A. Smirnov, S. Petrov, Kapitza Institute for Physical Problems, Moscow, Russia,
A. Shapiro, Shubnikov Institute for Crystallography, Moscow, Russia

Leon Balents (KITP), **H. Katsura** (Gakushuin U, Tokyo), **M. Kohno** (NIMS, Tsukuba)
Jianmin Sun (U Indiana), **Suhas Gangadharaiah** (U Basel)



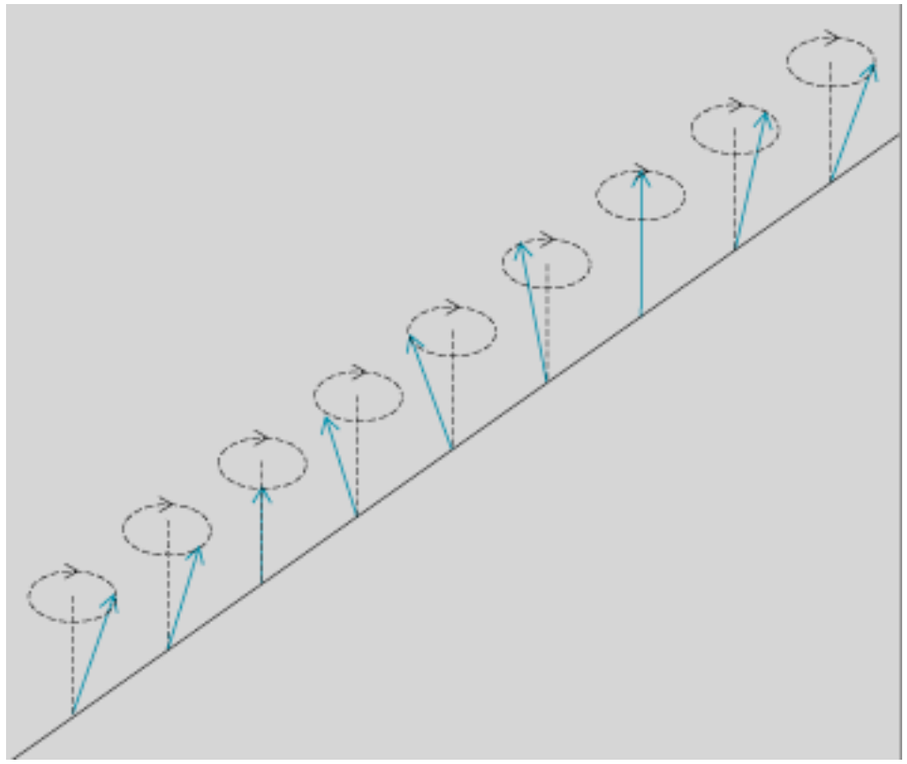
arxiv:1101.5275
Phys. Rev. B 82, 014421 (2010)
Phys. Rev. B 78, 054436 (2008)
Nature Physics 3, 790 (2007)
Phys. Rev. Lett. 98, 077205 (2007)

Spin Waves 2011, St. Petersburg

Outline

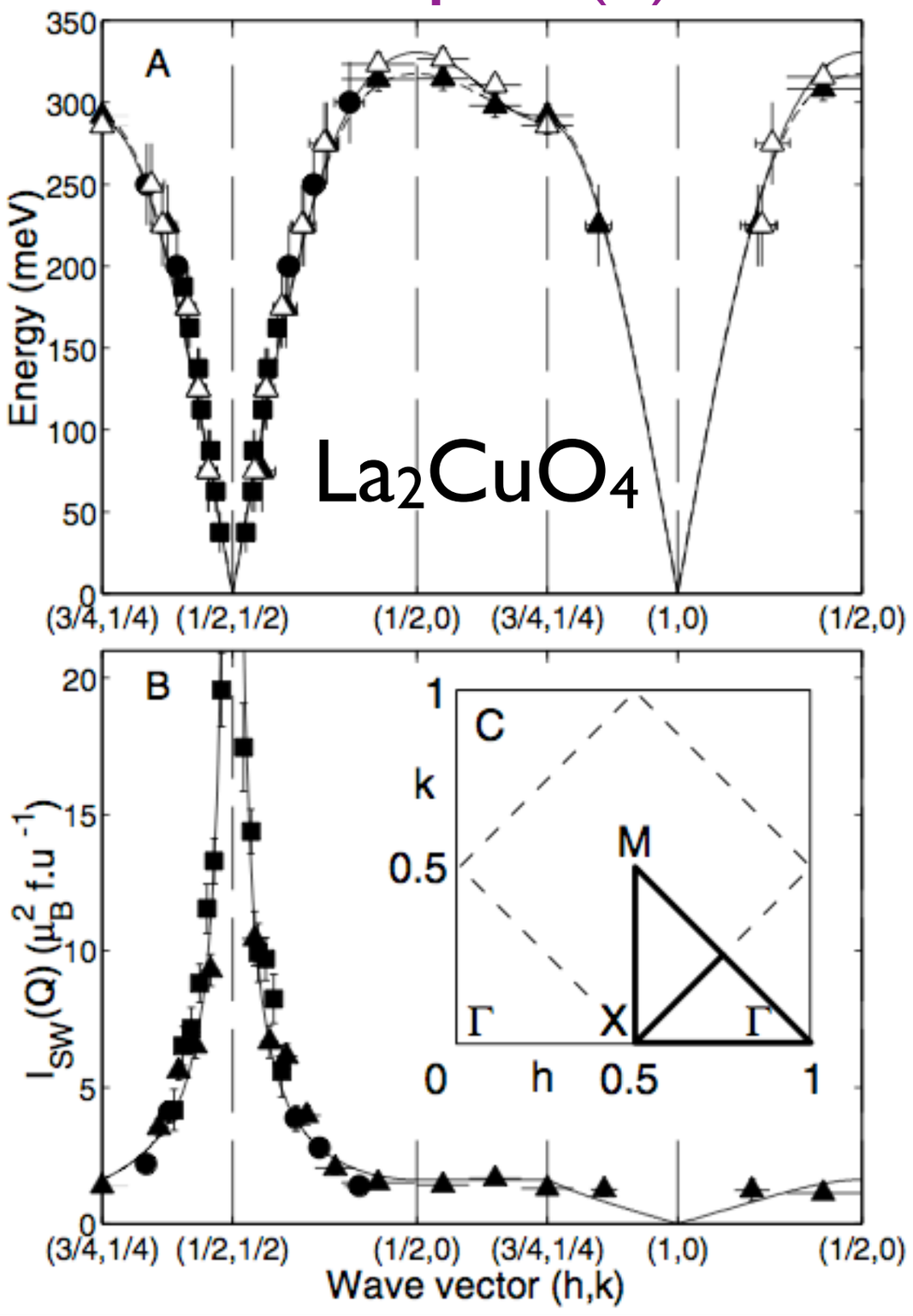
- Spin waves and *spinons*
- Experimental observations of spinons
  - ➡ neutron scattering, thermal conductivity, $2k_F$ oscillations, **ESR**
- Cs_2CuCl_4: spinon continuum and ESR
- ESR in the presence of uniform DM interaction
  - ESR in 2d electron gas with Rashba SOI
  - ESR in spin liquids with spinon Fermi surface
- Conclusions

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.

magnon is a boson

(neglecting finite dimension of the Hilbert space for finite S)

Coldea et al PRL 2001

But the *history* is more complicated

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."

But the *history* is more complicated

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."

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

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.

I. Pomeranchuk, JETP 1940

But the *history* is more complicated

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."

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

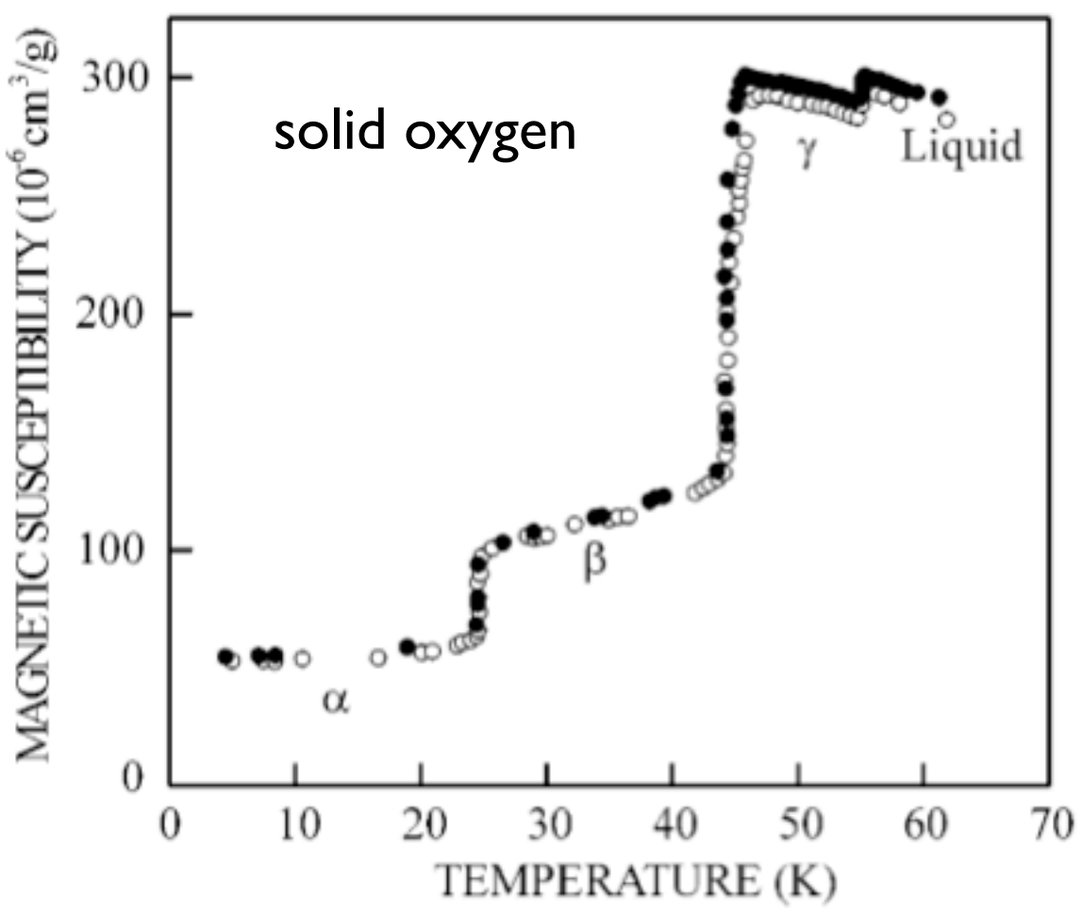
THE THERMAL CONDUCTIVITY OF THE PARAMAGNETIC DIELECT
LOW TEMPERATURES

By I. POMERANCHUK

(Received October 25, 1940)

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

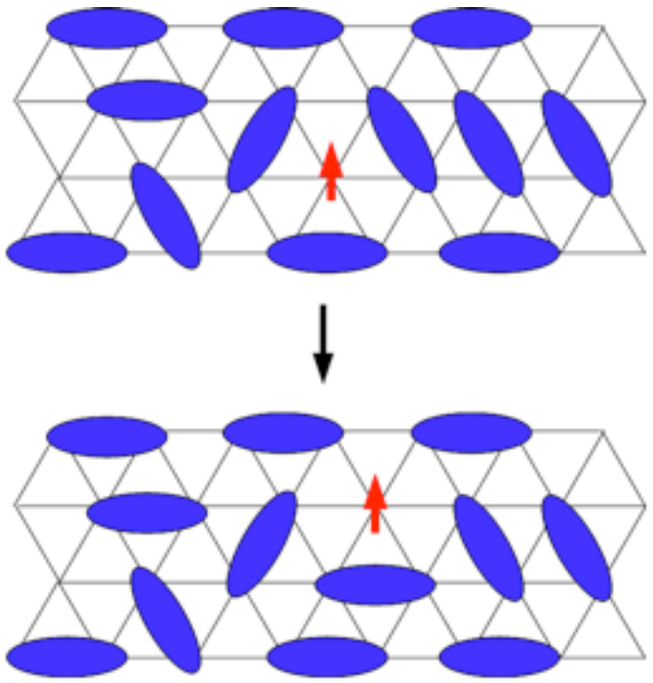
I. Pomeranchuk, JETP 1940



30+ years later

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

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



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.

and the arguments are still evolving …

Spinons are natural in d=1

Bethe's solution: 1933
identification of spinons: 1981

doi:10.1016/0375-9601(81)90335-2 | How to Cite or Link Using DOI
Copyright © 1981 Published by Elsevier Science B.V. All rights reserved.
Permissions & Reprints

What is the spin of a spin wave?

L. D. Faddeev and L. A. Takhtajan

Leningrad Branch of the Steklov Mathematical Institute, Leningrad, USSR

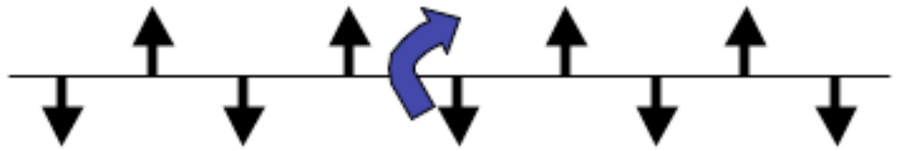
Received 15 July 1981. Available online 16 September 2002.

Abstract

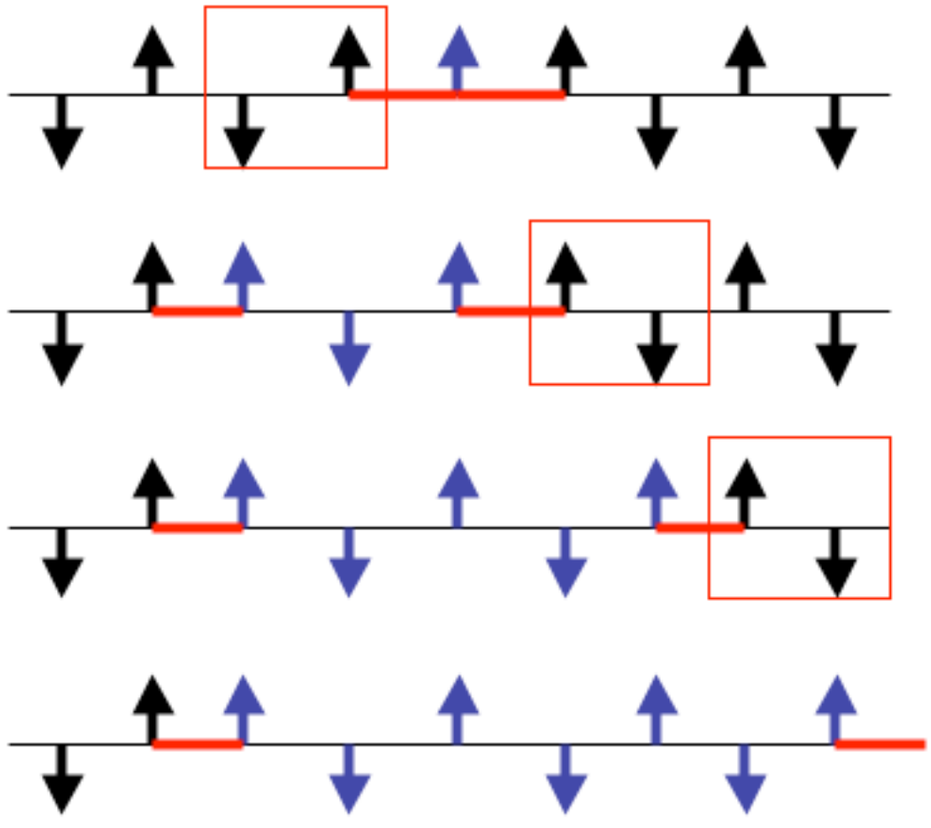
We argue that the spin of a spin wave in the Heisenberg antiferromagnetic chain of spins $\frac{1}{2}$ is equal to $\frac{1}{2}$ rather than 1 as is generally considered to be true.

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

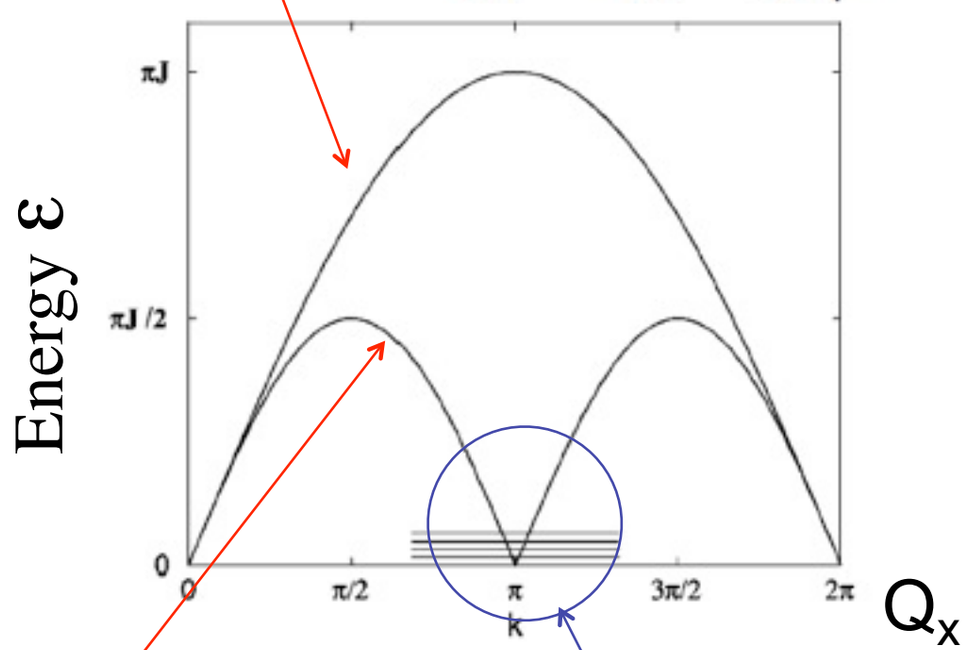
S=1 excitation $\begin{cases} \varepsilon = \omega(k_{x1}) + \omega(k_{x2}) \\ Q_x = k_{x1} + k_{x2} \end{cases}$

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

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

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

Upper boundary

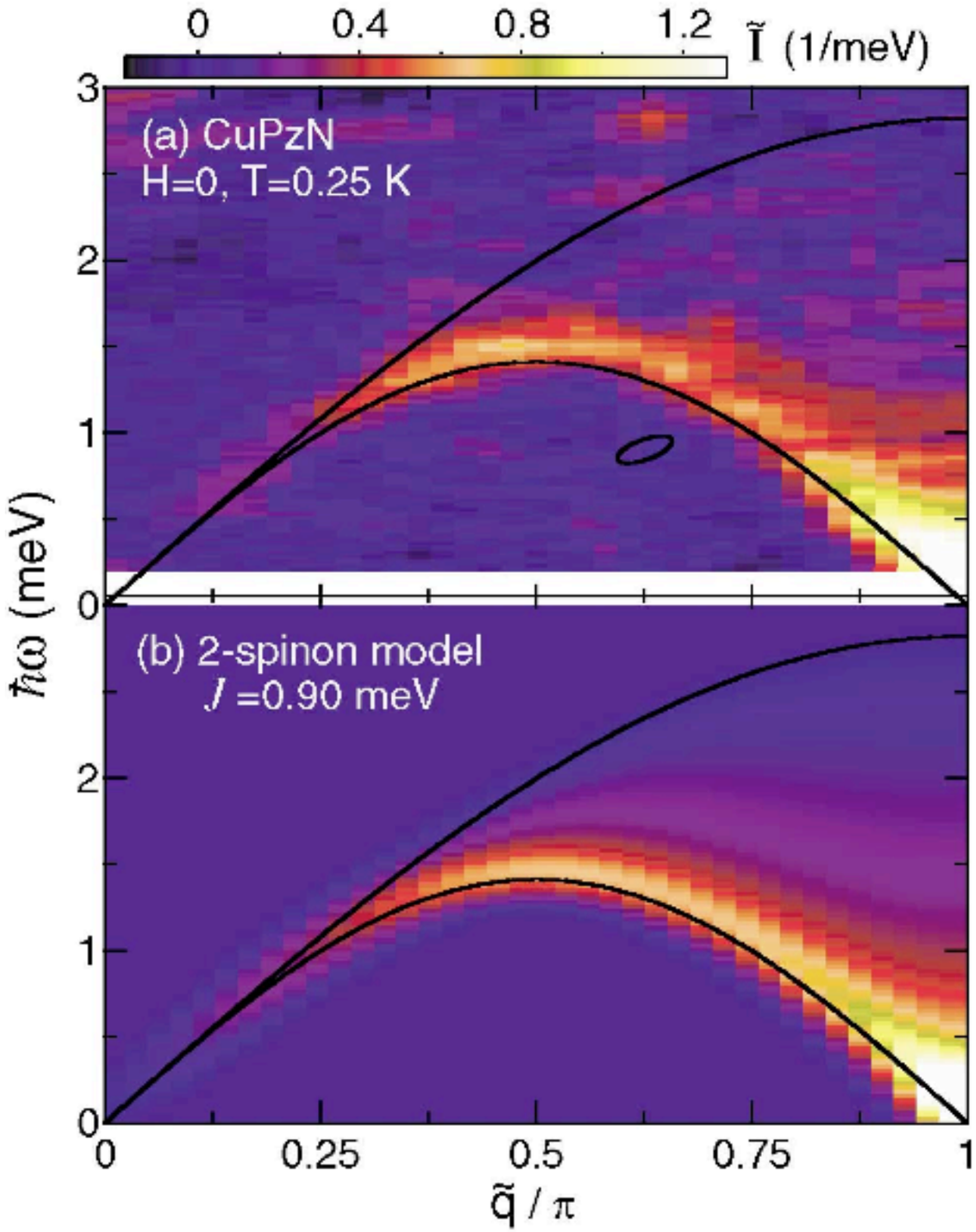


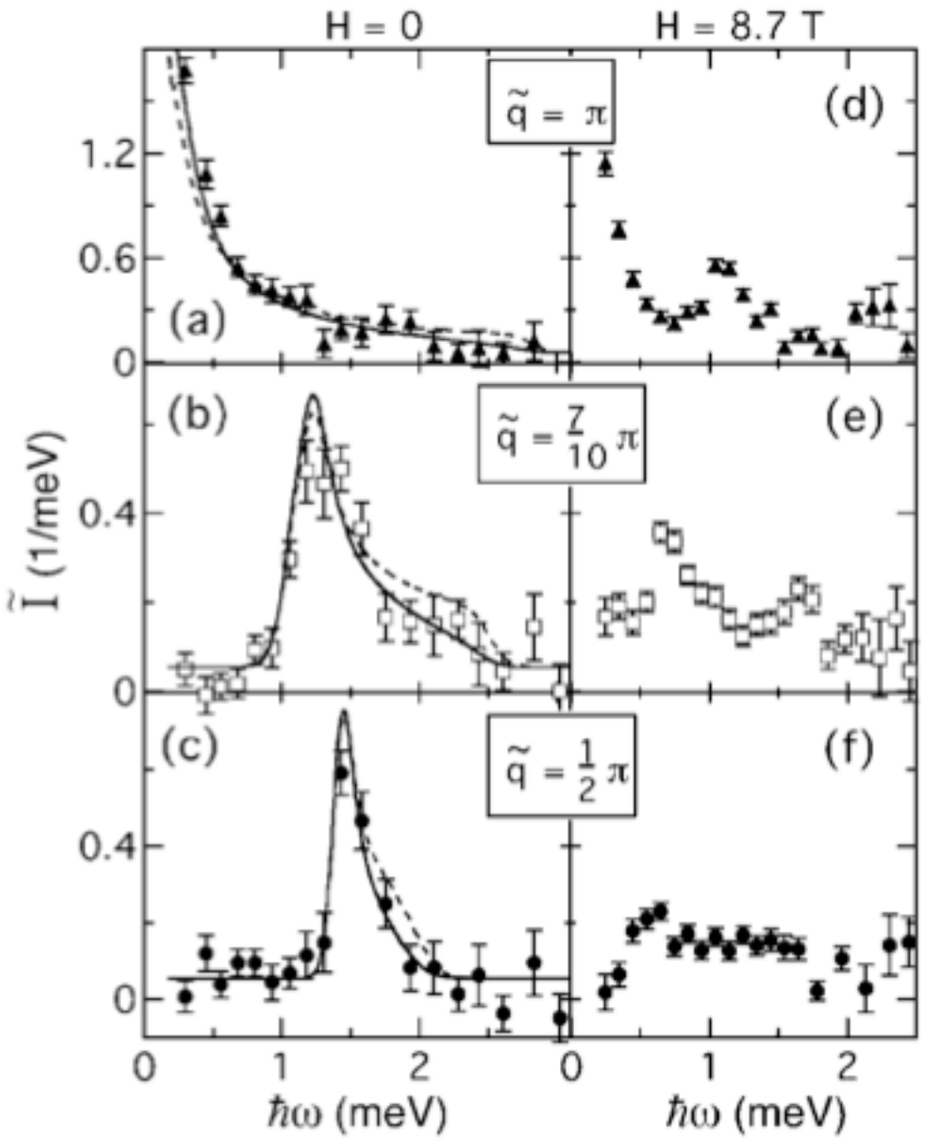
Lower boundary

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

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

Highly 1d antiferromagnet CuPzN



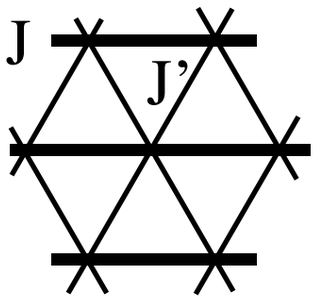


Stone et al, PRL 2003

$J'/J < 10^{-4}$

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]

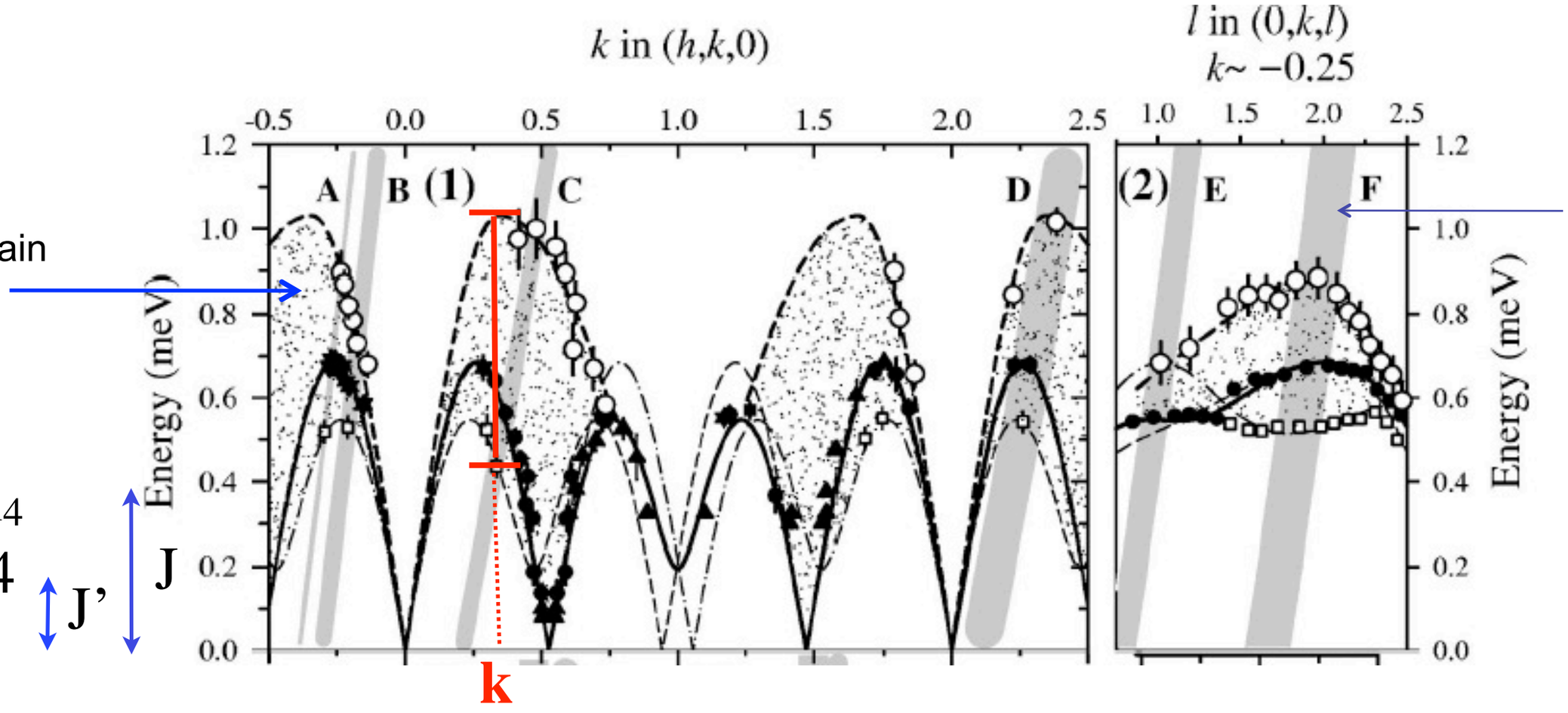


PHYSICAL REVIEW B 68, 134424 (2003)

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



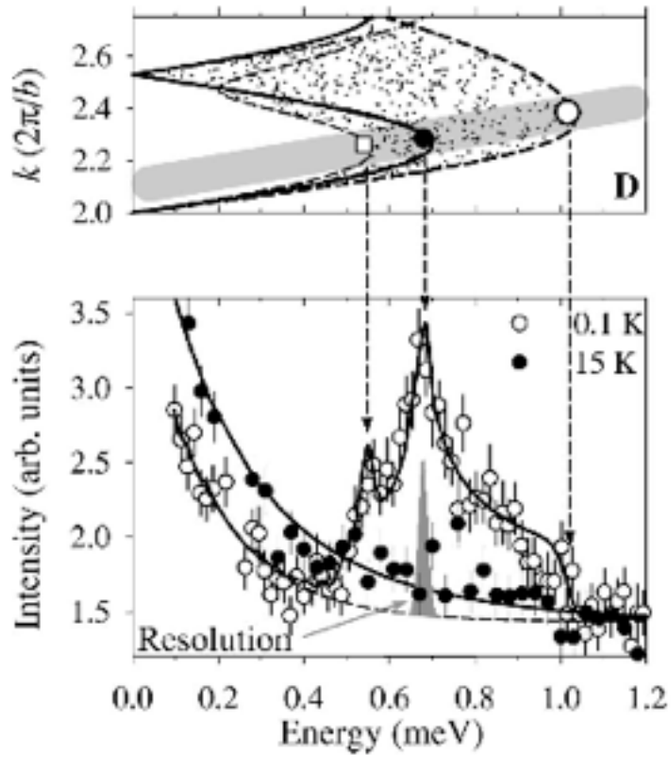
Cs_2CuCl_4
J'/J=0.34

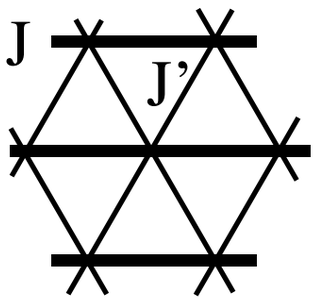
J' J

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

Experimental Realization of a 2D Fractional Quantum S

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



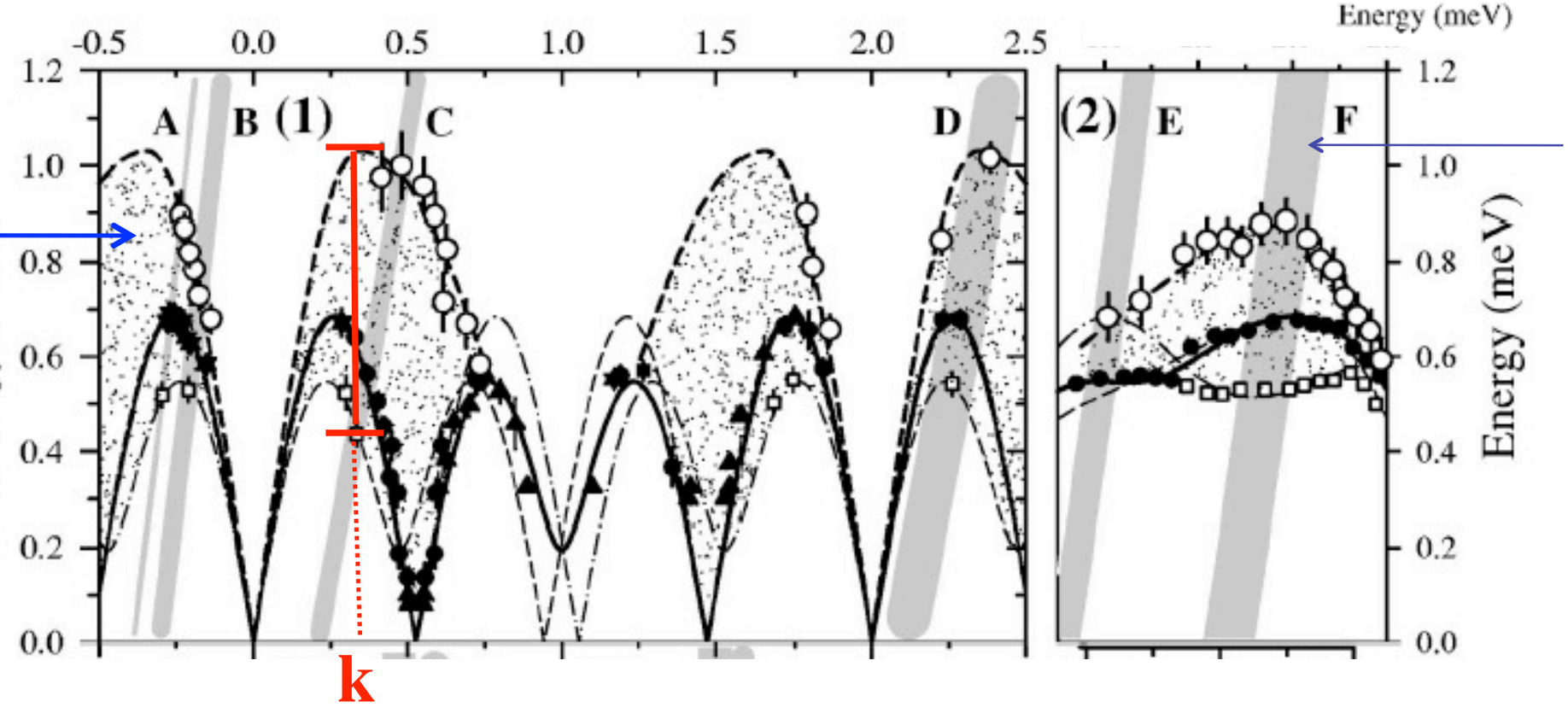


PHYSICAL REVIEW B 68, 134424 (2003)

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]

k in $(h,k,0)$



Along the chain

transverse to chain

Cs_2CuCl_4

$J'/J=0.34$

J'

J

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

Effective Schrödinger equation in two-spinon basis

Kohno, OS, Balents: Nat. Phys. 2007

- Study two spinon subspace

$$|k_x, k_y; \epsilon\rangle = \sum_y e^{ik_y y} |k_x, \epsilon\rangle_y \otimes_{y' \neq y} |0\rangle_{y'}$$

(two spinons on chain **y** with $S^z_{tot} = +1$)

  - Momentum conservation: 1d Schrödinger equation in ε space (**k** = (k_x, k_y))

$$\epsilon \psi_{\mathbf{k}}(\epsilon) + \int d\tilde{\epsilon} D_{k_x}(\tilde{\epsilon})\, J'(\mathbf{k}) A^*_{k_x}(\epsilon) A_{k_x}(\tilde{\epsilon})\, \psi_{\mathbf{k}}(\tilde{\epsilon}) = E \psi_{\mathbf{k}}(\epsilon)$$

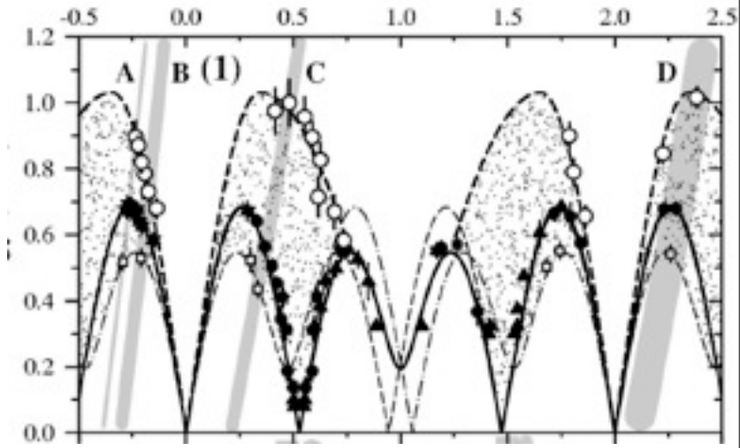
- Crucial matrix elements known exactly

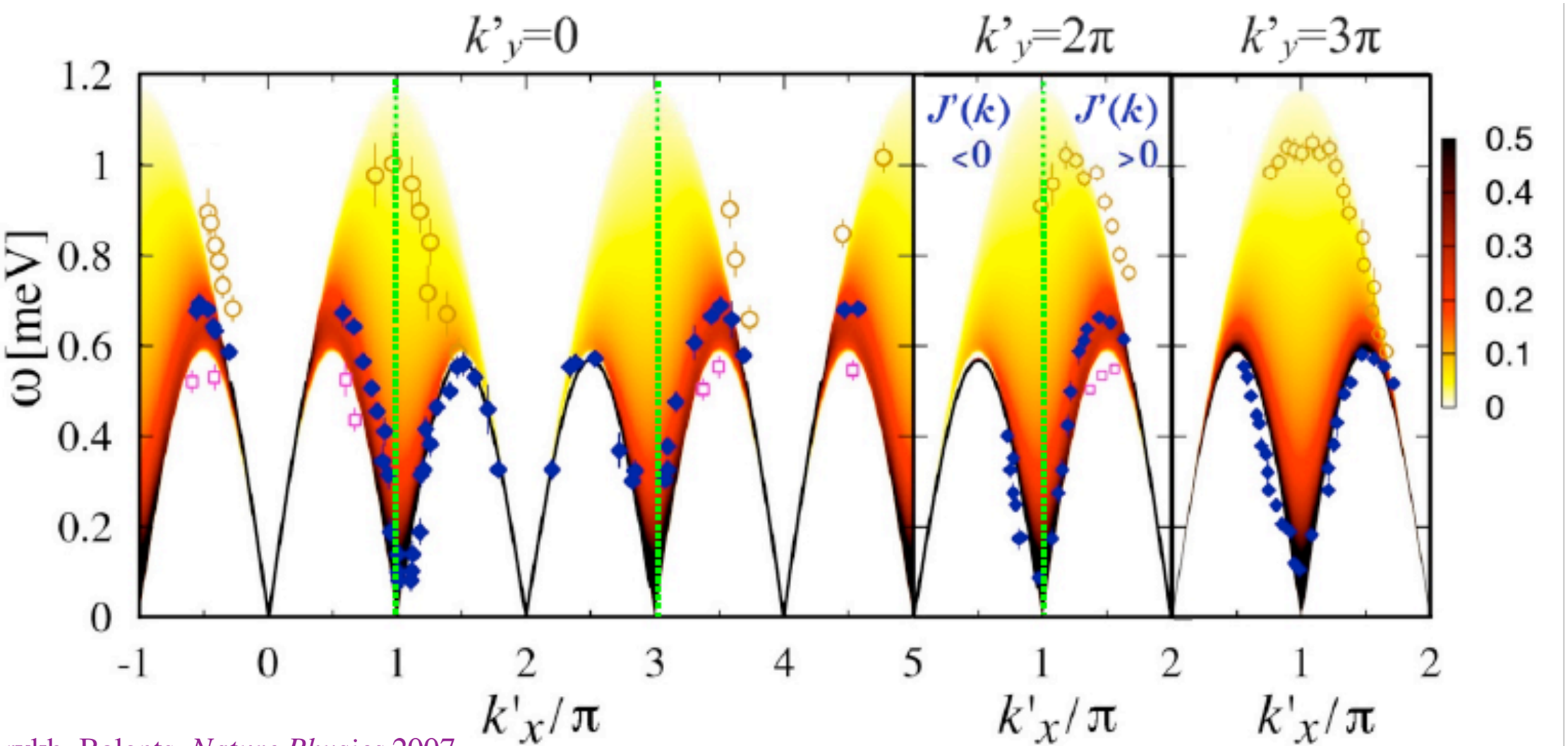
$$A_{k_x}(\epsilon) \equiv \frac{1}{\sqrt{2}} \langle 0 | S^-_{-k_x, y} | k_x, \epsilon \rangle_y$$

Bougourzi *et al*, 1996

- Calculate dynamic spin structure factor

Dimensional reduction due to frustration:
spinons in Cs_2CuCl_4 are of 1d origin,
propagate between chains
by forming S=1 triplon pairs





Kohno, Starykh, Balents, *Nature Physics* 2007

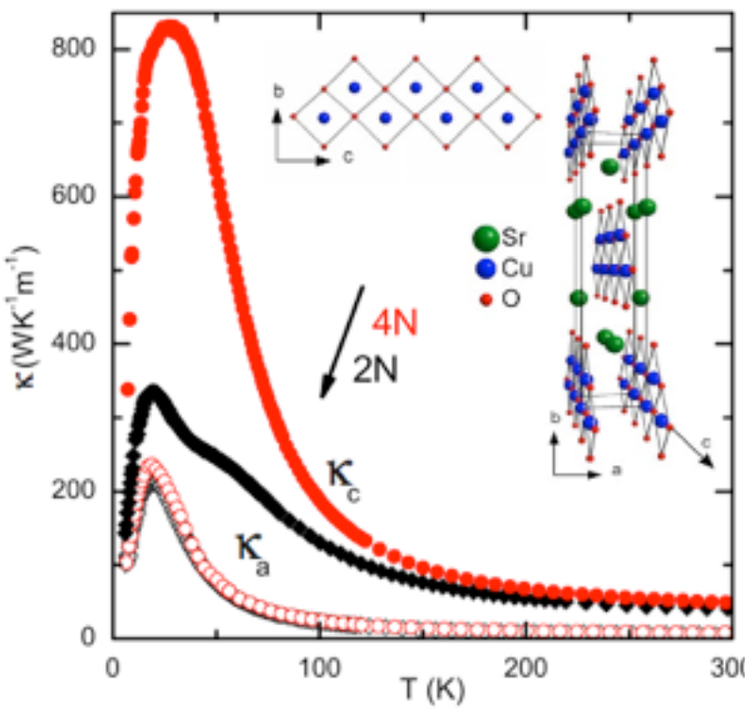
- Vertical green lines: $J'(k)=4J' \cos[k'_x/2] \cos[k'_y/2] = 0$.

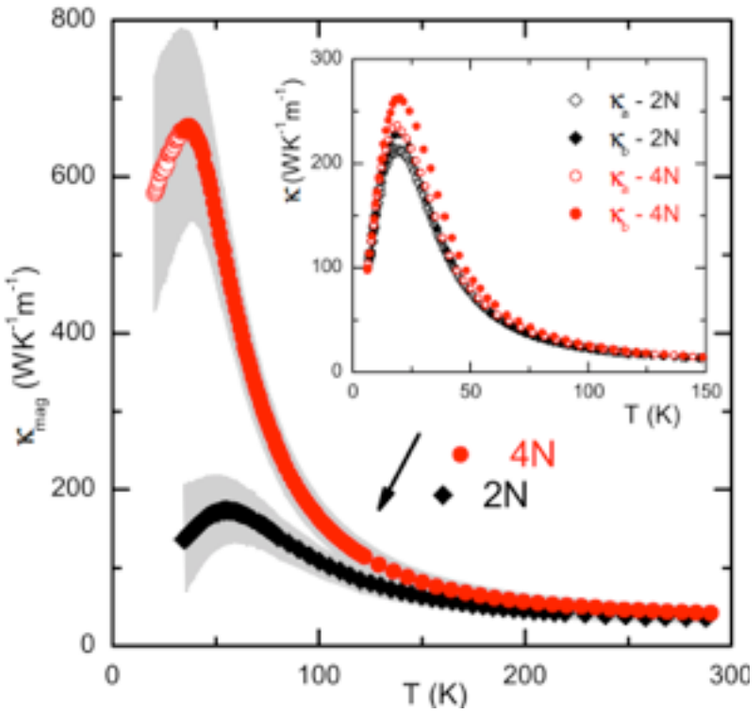
How else can we probe/see spinons?

PHYSICAL REVIEW B **81**, 020405(R) (2010)

Ballistic heat transport of quantum spin excitations as seen in $SrCuO_2$

N. Hlubek,[1] P. Ribeiro,[1] R. Saint-Martin,[2] A. Revcolevschi,[2] G. Roth,[3] G. Behr,[1] B. Büchner,[1] and C. Hess[1]
[1]*IFW-Dresden, Institute for Solid State Research, P.O. Box 270116, D-01171 Dresden, Germany*
[2]*Laboratoire de Physico-Chimie de L'Etat Solide, ICMMO, UMR 8182, Université Paris-Sud, 91405 Orsay, France*
[3]*Institut für Kristallographie der RWTH, D-52056 Aachen, Germany*
(Received 11 November 2009; published 11 January 2010)





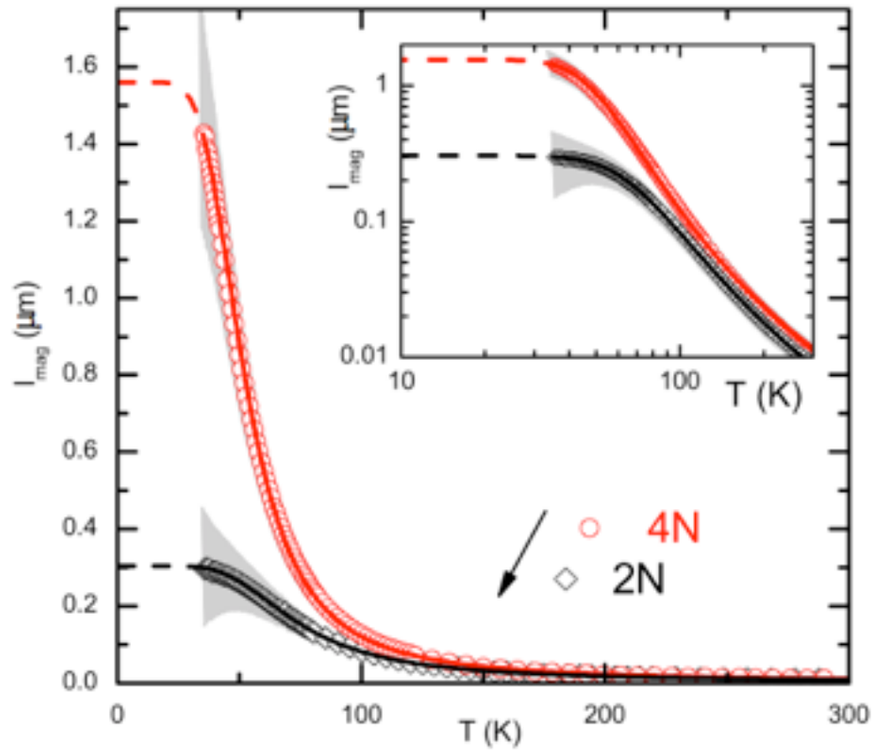


FIG. 3. (Color online) Magnetic mean-free paths of $SrCuO_2$ for different purities. The solid lines were calculated according to Eq. (2). The shaded area illustrates the uncertainty from the estimation of the phononic background.

mean-free path ~1 micron !

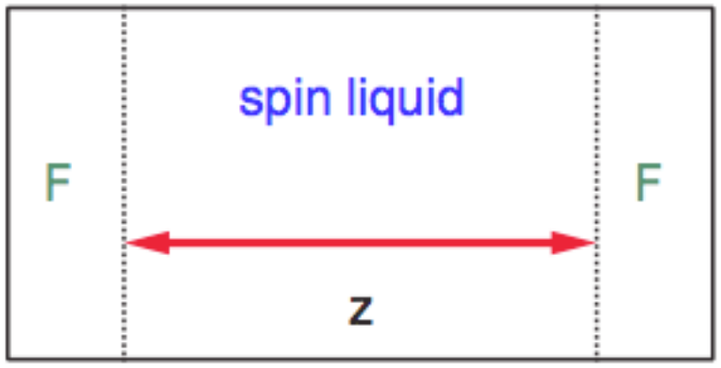
Friedel oscillations due to spinon Fermi surface

PRL 102, 067204 (2009) PHYSICAL REVIEW LETTERS week ending 13 FEBRUARY 2009

How to Measure a Spinon Fermi Surface

M. R. Norman and T. Micklitz
Materials Science Division, Argonne National Laboratory, Argonne, Illinois 60439, USA
(Received 15 September 2008; revised manuscript received 30 October 2008; published 12 February 2009)

RKKY like coupling mediated by spinons



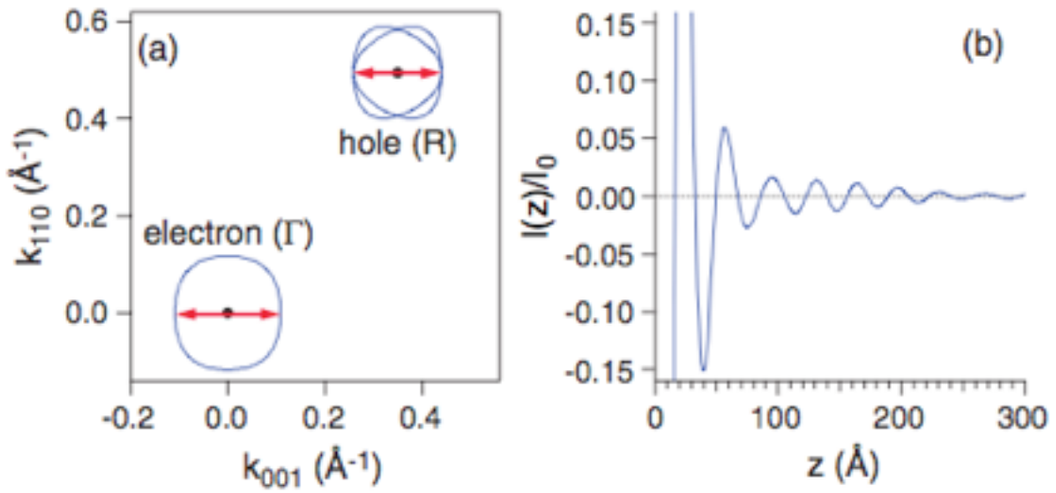


FIG. 3 (color online). (a) Spinon Fermi surface for $Na_4Ir_3O_8$ ($a = 8.985$ Å). Spanning vectors are indicated by arrows. (b) Calculated oscillatory response from Eq. (12).

- Charge Friedel oscillations in a Mott insulator, Mross and Senthil, arxiv:1007.2413

ESR - electron spin resonance

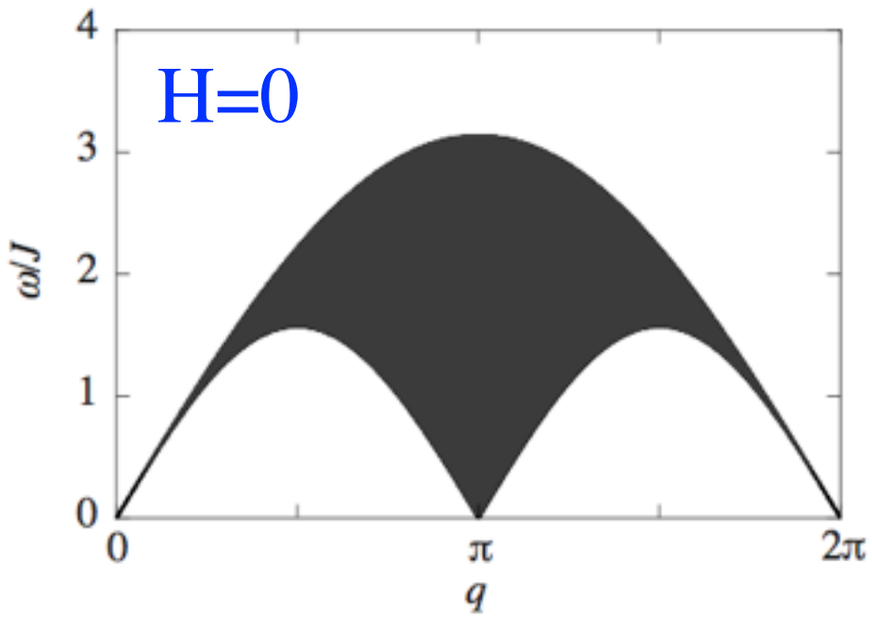
- Simple (in principle) and sensitive probe of magnetic *anisotropies* (and, also, **q=0** probe: $S = \Sigma_r S_r$)

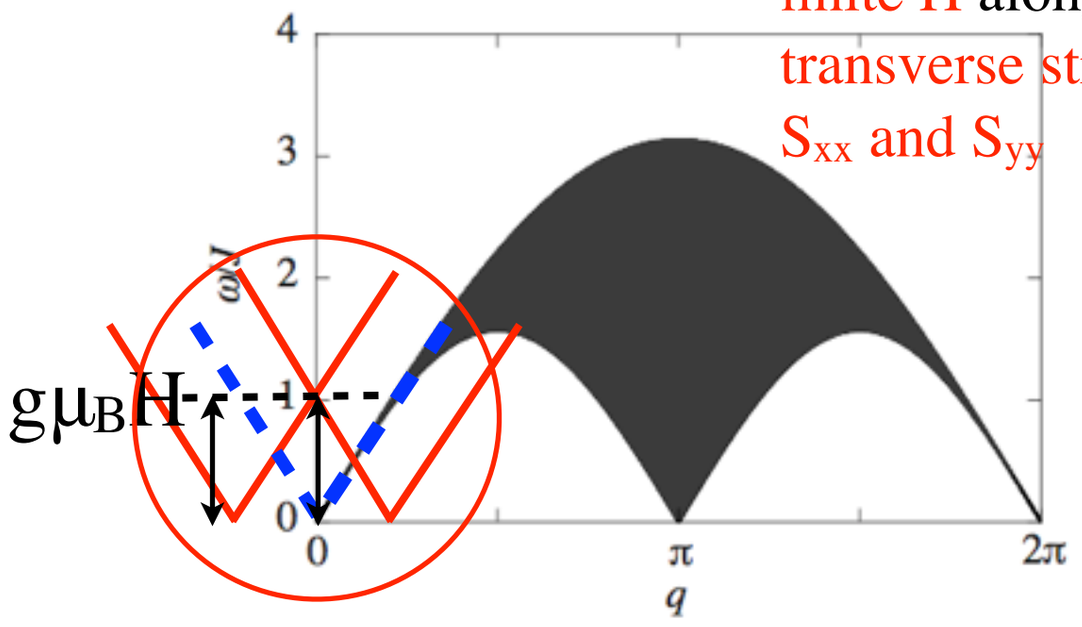
$$I(h,\omega) = \frac{\omega}{4L}\int dt e^{i\omega t}\langle [S^+(t), S^-]\rangle$$

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

- For SU(2) invariant chain in paramagnetic phase

  $I(H,\omega) \sim \delta(\omega\text{-}H)\ m(H)$ [Kohn's Th] Oshikawa, Affleck PRB 2002

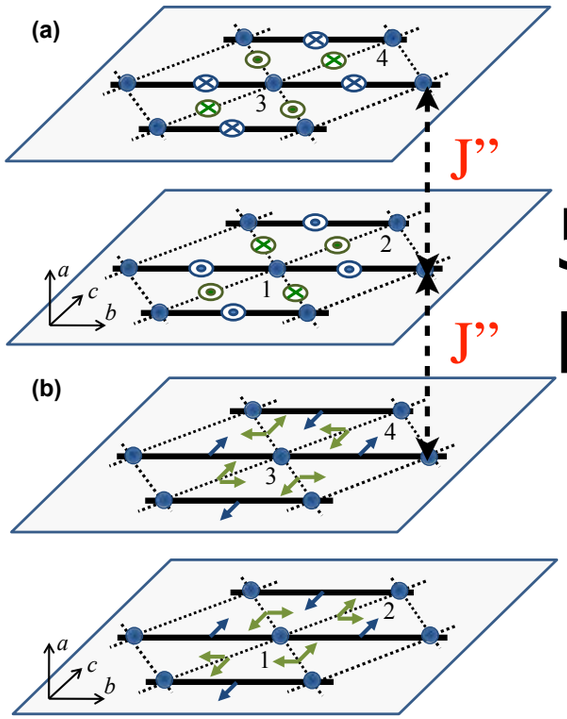




Main anisotropy in Cs_2CuCl_4: asymmetric Dzyaloshinskii-Moriya interaction

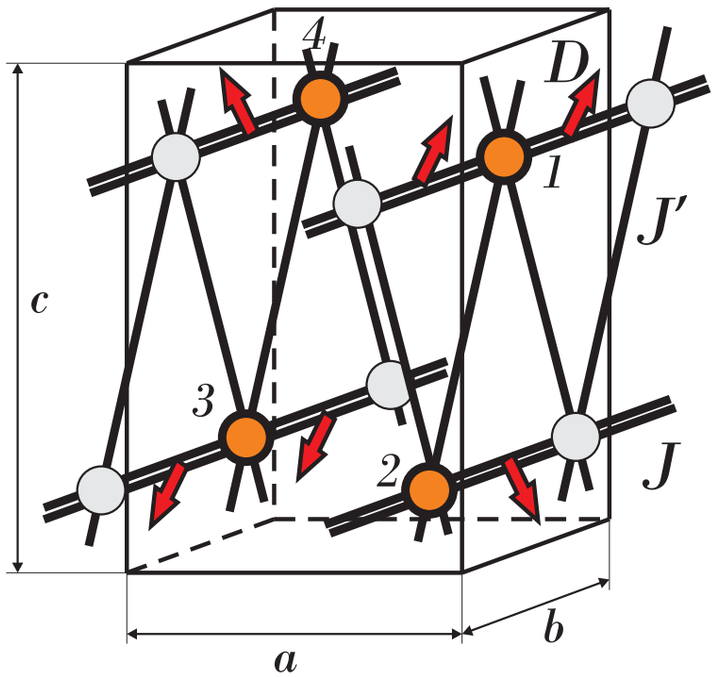
$$\vec{D}_{ij} \cdot \vec{S}_i \times \vec{S}_j$$

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



5 different DM terms allowed

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



focus on the in-chain DM

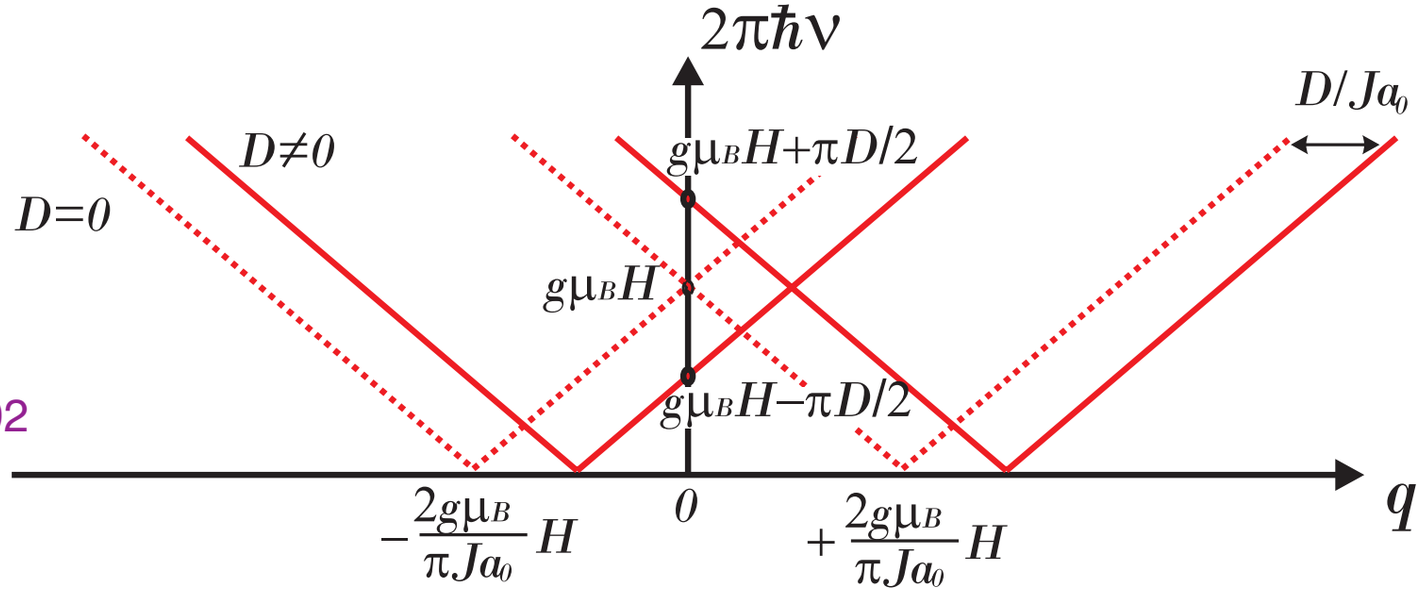
Theory I: H along DM axis

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

chain — uniform DM along the chain — magnetic field

- Unitary rotation $S^+(x) \rightarrow S^+(x)\, e^{i D x/J}$ removes DM term from the Hamiltonian (to D^2 order)
  - This ***boosts*** momentum to $D/(J a_0)$

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



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

D=0 picture
Oshikawa, Affleck 2002

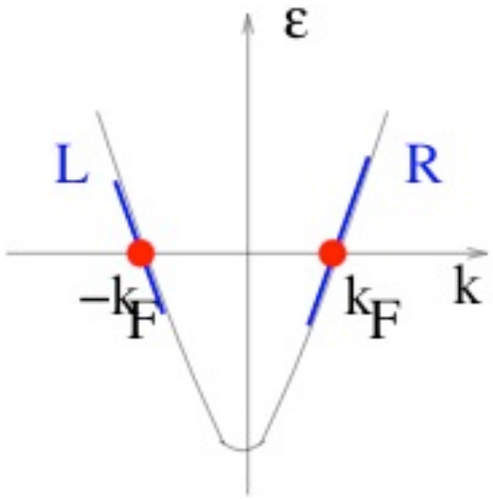
Chiral probe: **ESR probes *right*- and *left*-moving modes (spinons) independently**

Theory 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*

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

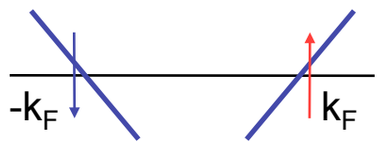
- *$2k_F$ (= π/a) fluctuations: charge density wave ε , spin density wave N*

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

$\chi_{1d}(q)$

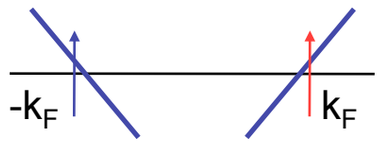
1/q

Staggered Dimerization

$\varepsilon = (-1)^x S_x S_{x+a}$

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

ΔS=0



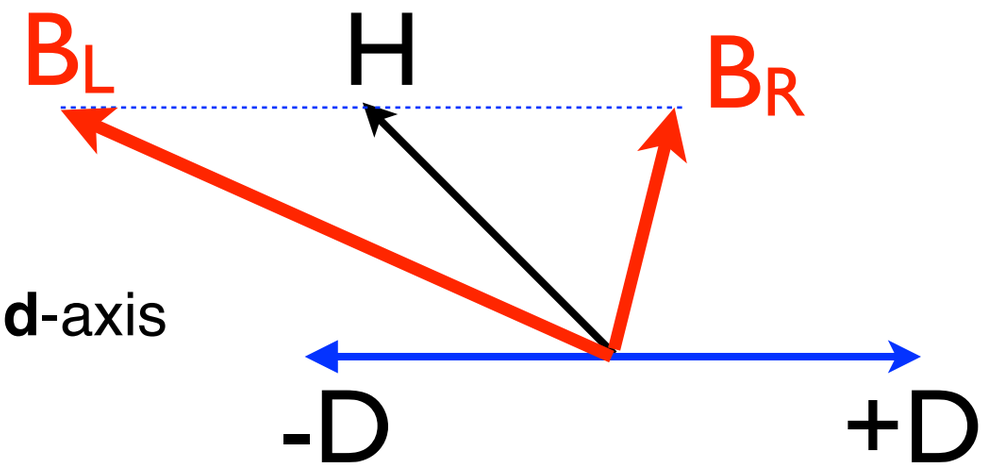
1/q

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

Theory II: arbitrary orientation (cont'd)

$$H = \boxed{\frac{2\pi v}{3}[(\vec{M}_R)^2 + (\vec{M}_L)^2]} \quad \boxed{-\frac{vD}{J}[M_R^d - M_L^d]} \quad -g\mu_B H[M_R^z + M_L^z]$$

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} ll b** is most effective.

Gangadharaiah, Sun, OS, PRB 2008

Temperature regimes

Paramagnetic

individual spins

universal quasi-classical regime

spin-correlated (spin liquid)

well-developed correlations along chains;
but little correlations between chains

ordered phase

strongly coupled chains;
2d (or 3d) description

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

J S

correlated spins (high T field theory)

$T_0 \sim J\, e^{-2\pi S}$

Haldane scale does exist for S=1/2 chains: different response for S=1 and S=1/2 chains below this temperature

spinons

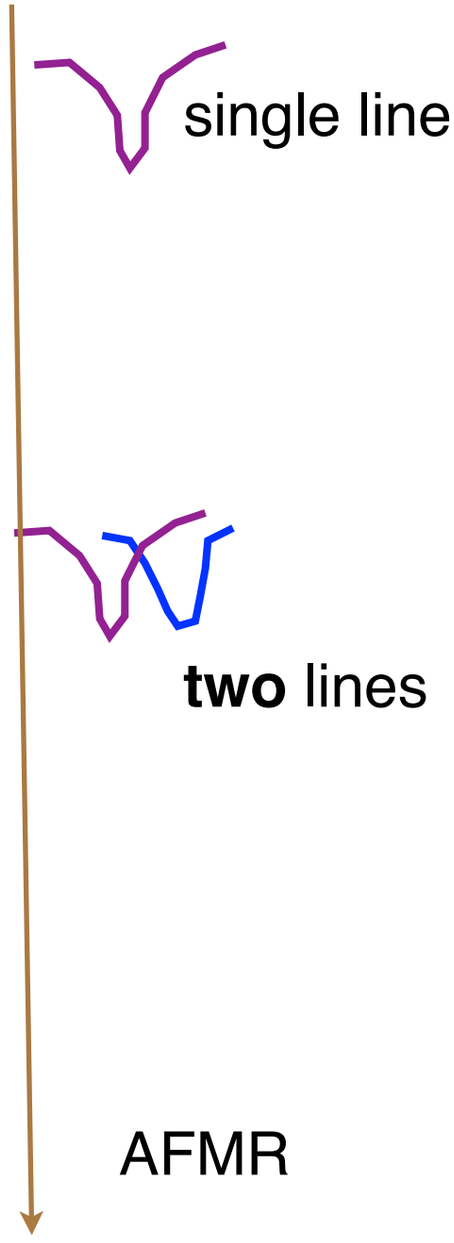
(low T field theory)

$T_N = 0.6$ K

spin waves

(at low energy)

ESR



Povarov et al, 2011

Temperature regimes

Paramagnetic

individual spins

universal quasi-classical regime

J S

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

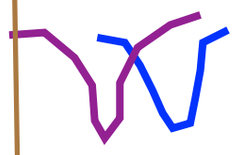
spin waves

(at low energy)

ESR



single line



two lines

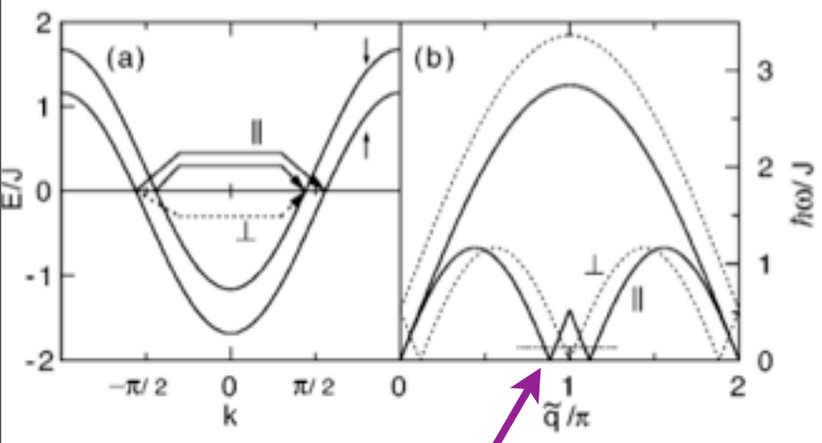
AFMR

Povarov et al, 2011

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

Another (more frequent) geometry: **staggered** DM

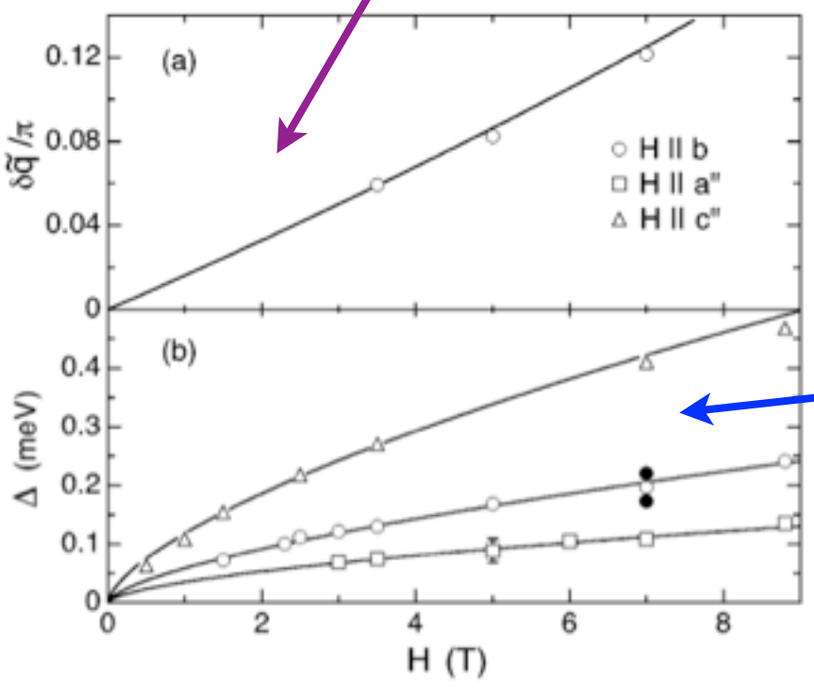
VOLUME 79, NUMBER 9 PHYSICAL REVIEW LETTERS 1 SEPTEMBER 1997



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

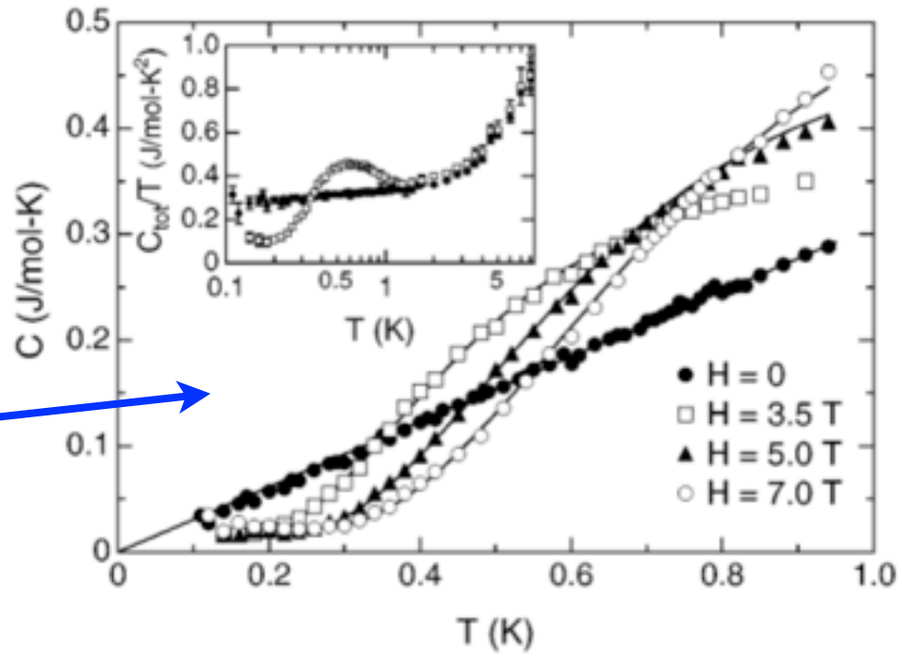
D. C. Dender,[1] P. R. Hammar,[1] Daniel H. Reich,[1] C. Broholm,[1,2] and G. Aeppli[3]
[1]*Department of Physics and Astronomy, The Johns Hopkins University, Baltimore, Maryland 21218*
[2]*National Institute of Standards and Technology, Gaithersburg, Maryland 20899*
[3]*NEC Research Institute, 4 Independence Way, Princeton, New Jersey 08540*
(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

$\vec{D}\cdot\vec{S}_n\times\vec{S}_{n+1}$	$(-1)^n\vec{D}\cdot\vec{S}_n\times\vec{S}_{n+1}$
h=0: free spinons	free spinons
finite h: free spinons	confined spinons
but subject to varying with momentum magnetic field	generate strongly relevant transverse magnetic field that binds spinons together $(-1)^n\frac{\vec{D}\times\vec{h}}{2J}\cdot\vec{S}_n$
ESR: generically two lines	single line
✓ splitting ✓ shift ✓ width (?)	✓ shift ✓ width

Analogy with Rashba S-O interaction in 2d electron gas

PRL 98, 187203 (2007) PHYSICAL REVIEW LETTERS week ending 4 MAY 2007

g-Factor Tuning and Manipulation of Spins by an Electric Current

Zbyslaw Wilamowski,[1,2] Hans Malissa,[1] Friedrich Schäffler,[1] and Wolfgang Jantsch[1]
[1]*Institut für Halbleiter- und Festkörperphysik, Johannes Kepler Universität, A-4040 Linz, Austria*
[2]*Institute of Physics, Polish Academy of Sciences, Warsaw, and Faculty of Mathematics and Computer Sciences, UWM Olsztyn, Poland*
(Received 2 October 2006; published 3 May 2007)

We investigate the Zeeman splitting of the two-dimensional electron gas in an asymmetric silicon quantum well, performing electron-spin-resonance (ESR) experiments. Applying a small dc current we observe a shift in the resonance field due to the additional current-induced Bychkov-Rashba type of spin-orbit field. We also show that a high frequency current may induce electric dipole spin resonance very efficiently. We identify different contributions to this type of ESR signal.

Original suggestion:
Kalevich, Korenev
JETP Lett 52, 230 (1990)

$$\alpha_R(p_x\sigma_y - p_y\sigma_x) \longrightarrow \alpha_R j_x\,\sigma_y$$

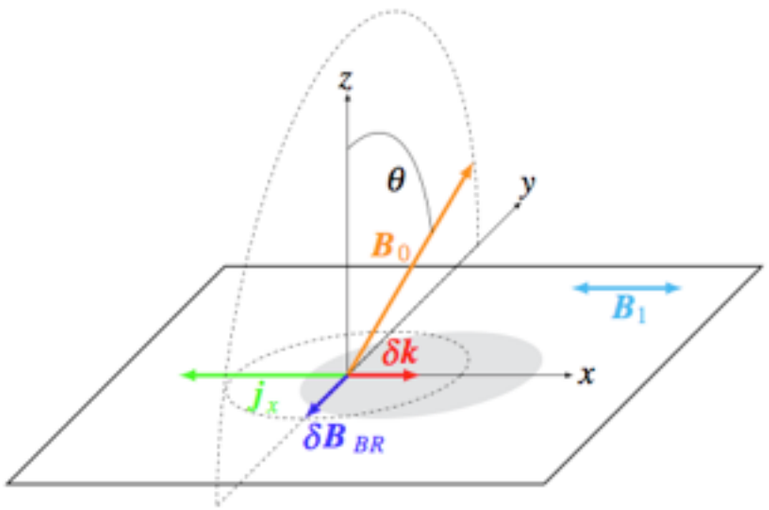


FIG. 1 (color online). A current j_x passes through a 2DEG (x-y plane). The Fermi circle (momentum distribution, dashed for $j_x = 0$, gray for $j_x \neq 0$), shifts by an amount $\hbar\delta\mathbf{k}$. Within this approximation, each electron experiences a Bychkov-Rashba field $\delta B_{\mathrm{BR},y}$ in addition to that resulting from its momentum $\hbar\mathbf{k}$. The static field B_0 (not drawn to scale) is applied in the y-z plane to enable ESR measurements.

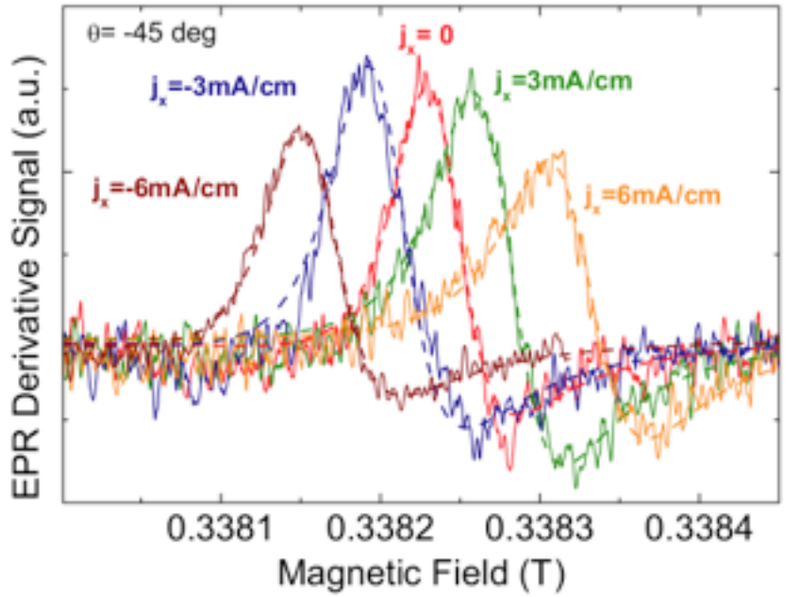


FIG. 2 (color online). ESR spectra of a 2DEG in a Si quantum well for various values of an electric current density passing a 3 mm wide sample. Measurements were performed with B_0 tilted by $\theta = -45°$ from the direction perpendicular to the sample plane at a microwave frequency of 9.4421 GHz.

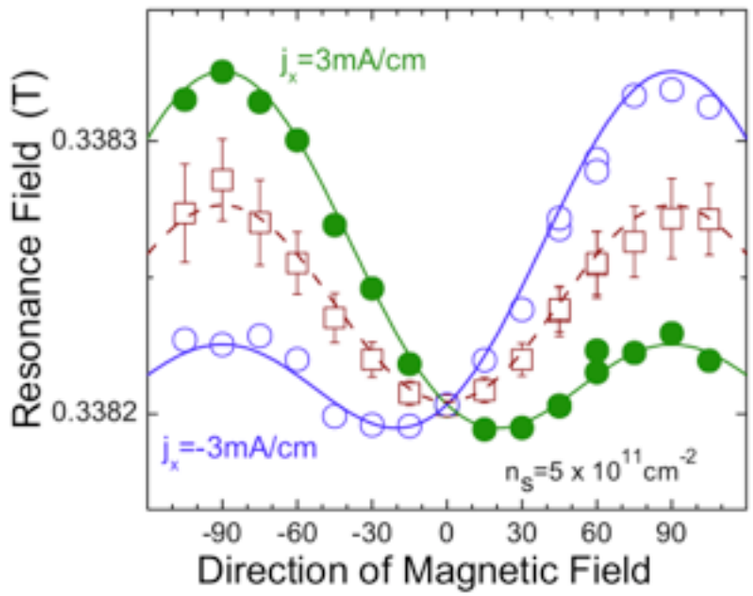


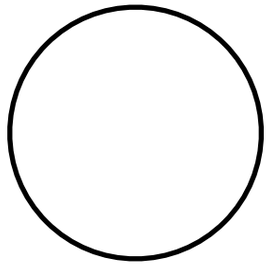
FIG. 4 (color online). Angular dependence of the ESR field for a current density $j = 0$ (squares) and ± 3 mA/cm (open and solid circles, respectively). The electron concentration is $n_S = 5 \times 10^{15}$ m^{-2} and the sample width 3 mm. Error bars correspond to 20% of the resonance linewidth.

Analogy with Rashba S-O interaction in 2d electron gas

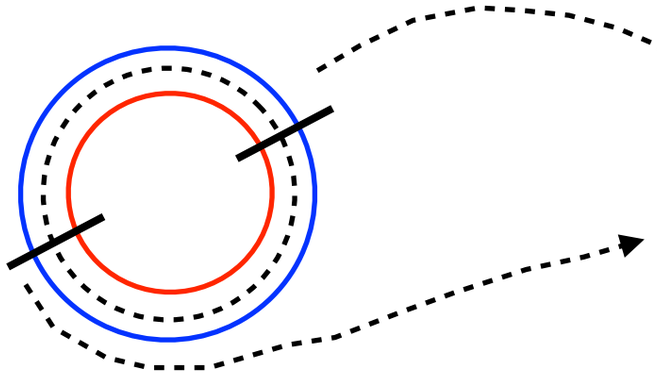
- 2d gas: dc current produces static Rashba field B_R (due to drift velocity) which adds to external B_0
- 1d AFM: uniform DM interaction produces **k**-dependent internal magnetic field which adds to external B_0
- But note that in 1d antiferromagnet the ***spinons*** are ***neutral fermions*** !
- Key question: can this approach be extended to two-dimensional spin liquids with spinon Fermi surface? (uniform RVB state of P. W. Anderson 1987)

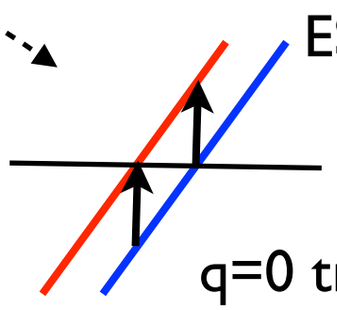
Tentative sketch for 2d (using Rashba model as an example)

no DM

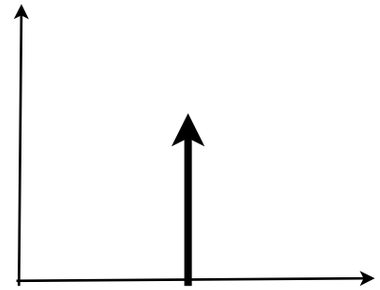


in-plane ***h***





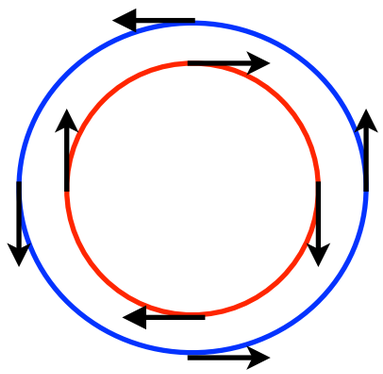
ESR signal



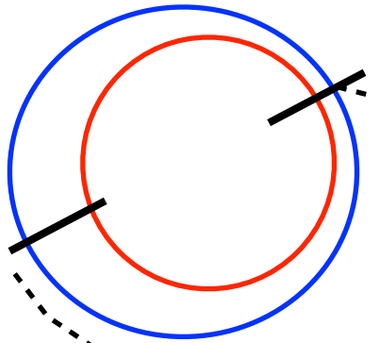
q=0 transitions at single ω = gμh

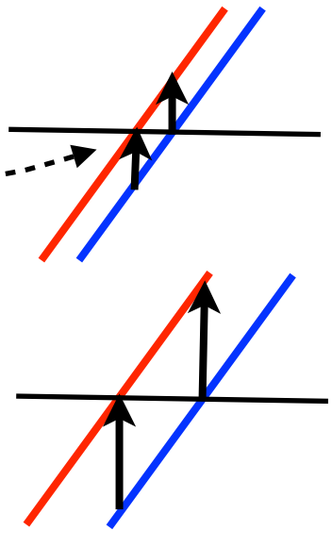
ω = gμh

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



in-plane ***h***





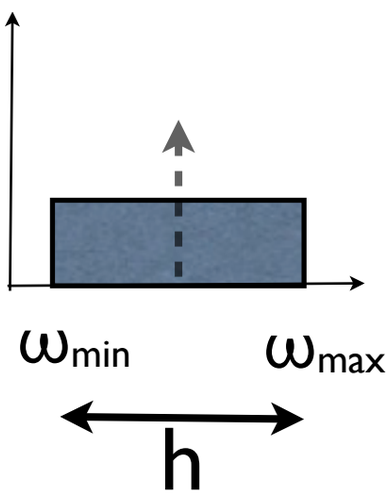
ω_{min}

ω_{max}

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

Raikh, Chen 1999

ESR signal



the width ~ h

line shape?

effect of gauge fluctuations?...

Conclusions

- Spinons *vs.* spin waves
- ESR as a novel probe of critical spinons (neutral fermions)
  - small momentum ~ D/J
  - allows to extract parameters of DM interaction
- Higher-dimensional extensions?!

"old" ESR probe
can access "new"
deconfined spinons
with a help from
DM interactions