Neutron scattering as a tool to study quantum magnetism

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Outline

- principles of (magnetic) neutron scattering
- spin waves in a Heisenberg ferromagnet
- spin waves in square-lattice AFM La$_2$CuO$_4$
- quantum renormalization, spinons and method to determine Hamiltonian – triangular AFM $S = 1/2$ Cs$_2$CuCl$_4$
- quantum phase transition in the Ising chain CoNb$_2$O$_6$ in transverse field
Neutron reactors
(Institute Laue Langevin)
- nuclear fission of Uranium

\[ k_i - k_f = Q \]
\[ E_i - E_f = E \]
Spallation neutron sources (ISIS, SNS …)

- “evaporation” when fast protons hit a heavy nucleus (Ta)

proton synchrotron accelerator $\sim 800\text{MeV}$

$\text{p}^+$

neutron guide

time-of-flight spectrometer

$\sim 40,000$ detector elements count simultaneously (time-stamp each arriving neutron)
Magnetic neutron diffraction

Neutrons have
- no charge
- spin-1/2 moment

Periodic magnetic order
=> magnetic Bragg peaks at $Q = \tau \pm q$

- Intensity $\sim |M_{\perp}(Q)|^2$

Fourier transform of magnetic moment density (perp to scattering wavevector $Q$)
Inelastic magnetic neutron scattering matrix element for transition

\[ \langle \lambda | S^\alpha(Q) | 0 \rangle \]

\[ S^\alpha(Q) = \frac{1}{\sqrt{N}} \sum_j e^{iQ \cdot R_j} S_j^\alpha \]

\[ S^{\alpha \alpha}(Q, \omega) = \sum \left| \langle \lambda | S^\alpha(Q) | 0 \rangle \right|^2 \delta(\hbar \omega + E_0 - E_\lambda) \]

\[ \frac{d^2 \sigma}{d\Omega dE'} \equiv \frac{k'}{k} Nr^2 \left| \frac{g}{2} F(Q) \right| e^{-2w(Q)} \sum_{\alpha \beta} \left( \delta_{\alpha \beta} - \hat{Q}_\alpha \hat{Q}_\beta \right) S^{\alpha \beta}(Q, \omega) \]

Fourier transform of magnetic e- density

polarization factor
Single crystals for inelastic neutron scattering

La$_2$CuO$_4$

Cs$_2$CuCl$_4$
(solution growth)

CoNb$_2$O$_6$
(mirror furnace growth)

7 single-crystal mount ~50 g
(flux growth)

2.5 cm

4 cm

floating-zone mirror furnace
Spin waves in a Heisenberg ferromagnet

\[ H = - \sum_{ij} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j \] if all \( J_{ij} > 0 \) \( T=0 \) ground state is \( \uparrow \uparrow \uparrow \uparrow \uparrow \uparrow \)

- neutrons flip over one spin \( S^- \uparrow \uparrow \uparrow \uparrow \uparrow \ldots = \uparrow \uparrow \uparrow \uparrow \uparrow \uparrow \)

**magnon energy**

\[ \omega(q) = -J(q) + J(0) + h \]

**exact result**

**Fourier transform of magnetic couplings**

\[ J(q) = \frac{1}{2} \sum_{ij} J_{ij} e^{iqr_{ij}} \]

**Zeeman energy**

**Wavevector \( (q) \)**

**Energy**

**Gap \( \Delta \)**

**Coherent propagation of spin-flip states**

(if Hamiltonian conservs \( S^z \))

**Dispersion images exchange Hamiltonian**

\[ |\varphi_q> = \frac{1}{\sqrt{N}} \sum_i e^{iqr_i} |\downarrow_i> \]
Neutron scattering by ferromagnetic magnons

incident neutron $k_i$
$S^z = -1/2$

scattered
$S^z = +1/2$

$k_f$

$k = k_i - k_f$

$E_i - E_f = \hbar \omega(k)$
Spin waves in the square-lattice anti-ferromagnet

Ground state has Neel order
(<S> reduced by quantum fluctuations)

$$S_i \cdot S_j = S_i^z S_j^z + \frac{1}{2} \left( S_i^+ S_j^- + S_i^- S_j^+ \right)$$

La$_2$CuO$_4$

- insulating parent of high-$T_C$ cuprates
- square-lattice of CuO$_2$ planes, Cu$^{2+}$ S=1/2

Spin wave excitations
(approximate eigenstates)

Magnetic Bragg peak (1/2,1/2,0)

Keimer at al, PRB (1992)
Neutron scattering experiments on La$_2$CuO$_4$

7 single crystal mount, ~50 g

RC et al, PRL 86, 5377 (2001)
Magnetic excitations in La$_2$CuO$_4$

Collect maps of magnetic scattering in the whole 2D Brillouin zone $(h,k)$ at increasing energies $E$

Spin-wave dispersion surface

RC et al, PRL 86, 5377 (2001)
Dispersion relations

La$_2$CuO$_4$, $T=293$ K

RC et al, PRL 86, 5377 (2001)
Dispersion relation and interactions

- dispersion shape is a direct fingerprint of the magnetic interactions

- “wiggle” in high-energy dispersion is evidence for a cyclic-exchange between the 4 spins on a plaquette in addition to the main exchange $J$

\[ \mathcal{H} = J \sum_{\langle i,j \rangle} S_i \cdot S_j + J_c \sum_{\langle i,j,k,l \rangle} \{(S_i \cdot S_j)(S_k \cdot S_l) + (S_i \cdot S_l)(S_k \cdot S_j) - (S_i \cdot S_k)(S_j \cdot S_l)\}, \] (1)

$J = 138 \text{ meV}$ \hspace{0.5cm} $J_c = 38 \text{ meV}$
Ring exchange in the Hubbard model

Expand up to 4 electron hops

\[ \mathcal{H} = J \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J' \sum_{\langle i,i' \rangle} \mathbf{S}_i \cdot \mathbf{S}_{i'} + J'' \sum_{\langle i,i'' \rangle} \mathbf{S}_i \cdot \mathbf{S}_{i''} + J_c \sum_{\langle i,j,k,l \rangle} \left\{ (\mathbf{S}_i \cdot \mathbf{S}_j)(\mathbf{S}_k \cdot \mathbf{S}_l) + (\mathbf{S}_i \cdot \mathbf{S}_l)(\mathbf{S}_k \cdot \mathbf{S}_j) \right\} - (\mathbf{S}_i \cdot \mathbf{S}_k)(\mathbf{S}_j \cdot \mathbf{S}_l) \]  

(1)

A.H. MacDonald (1990), Takahashi (1977)
Hubbard model parameters for \( \text{La}_2\text{CuO}_4 \)

- dispersions and intensities well described by linear spin-wave theory

\[ \begin{align*}
t &= 0.30(2) \text{ eV}, \quad U = 2.2 (4) \text{ eV} \\
U/t &= 7.3 \pm 1.3 \text{ (10 K)}
\end{align*} \]

Intensity renormalization factor
\[ Z_c = 0.51 \pm 0.13 \text{ (predicted 0.61)} \]

RC et al, PRL 86, 5377 (2001)
Magnetic excitations in $S = 1/2$ triangular lattice AFM Cs$_2$CuCl$_4$

- sharp spin-wave mode only very small weight
- dominant scattering continuum with strongly-dispersive boundaries

=> Quantum fluctuations very strong, spin-wave theory inadequate

Experimental method to determine Hamiltonian via spin waves in the fully-polarized state in high field

\[ \omega(k) = J(k) - J(0) + h \]

\[ J(k) = \frac{1}{2} \sum d J_d \exp(i k \cdot \delta) \]


- B=0
- B >> J
- \( J \) antiferromagnetic
- State with AF correlations
- Fully-polarized state
- Magnetic field
- \( B_C \)
- Bose condensation of magnons at \( B_C \)
- Exact eigenstate if Hamiltonian conserves \( S_z \)
- Dispersion is the Fourier transform of exchange couplings
- Gapped magnons
- Magnetically ordered, spin liquid etc.
Excitations in the saturated ferromagnetic phase at $B=12$ T

Fourier transform of couplings $J(q)

\[ J = 0.374(5) \text{ meV} \]

\[ J' = 0.128(5) \text{ meV} \]

\[ J'' = 0.017(2) \text{ meV} \]

interlayer coupling

\[ D_a = 0.020(0) \text{ meV} \]

DM anisotropy \( \perp \) bc plane

$B_c = 8.42$ T

$T = 60$ mK

Magnon condensation below critical field induces transverse order

Link magnetic order with magnon wavefunctions

2 condensates at +Q and -Q

The two condensates interact via the inter-layer couplings \( J'' \)

Asymmetric shape

\[ \langle S_b \rangle / \langle S_c \rangle \]

predicted asymmetry using magnon wavefunctions

Quantum renormalization of incommensurate ordering wavevector

$B = B_c$

$B < B_c$

Strong quantum renormalization

$\varepsilon_0 / \varepsilon_{cl} = 0.56$

$\epsilon = 0.0536(5)$

$B(T) || a$

Classical instability

Large-$S$

Spin-1/2

Collinear spins

$Q$

Mean-field result

Magnetic excitations at zero field: spin-waves fractionalize into pairs of $S=1/2$ spinons

$J'/J \sim 1/3, \; J=0.37$ meV

$S_i^+ S_j^- + S_j^- S_i^+$

Kohno, Starykh, Balents, Nat. Phys. (07)
anisotropic triangular lattice $S=1/2$ AFM $\text{Cs}_2\text{CuCl}_4$ has spiral order coexisting with strong quantum fluctuations

- renormalization of $Q$-vector and zone-boundary energy measured by quenching quantum fluctuations via field and revealing “classical” behaviour
- dominant continuum scattering (spin-waves fractionalize into pairs of spinons)
Ising magnets and phase transitions

- classical Ising model
  \[ H = - \sum_{i,j} J S_i^z S_j^z \]

- 2D model Onsager exact solution (1944)

add transverse field
- \( B S^x \)
- \( B (S^+ + S^-) / 2 \)

quantum fluctuations
quantum tunneling

Classical thermally-driven continuous phase transition

Quantum fluctuations driven continuous phase transition

\( T=0 \) “quantum melting of order”
An Ising ferromagnet in transverse field

\[ H = - \sum_i J S_i^z S_{i+1}^z - B S_i^x \]

- transverse field
  \[ -B S^x = -B (S^+ + S^-) / 2 \]

generates quantum fluctuations that “melt” the spontaneous magnetic order at \( B_C \sim J/2 \)

- continuous quantum phase transition

ordered moment

\[ \langle S^z \rangle \]

Magnetization

\[ \langle S^x \rangle \]
- what is **microscopic mechanism of transition**, can one observe the quantum fluctuations that drive transition?

- how **quasiparticles** evolve near critical point?

- what are the **fundamental symmetries** that govern physics of QCP?

- what are **finite-T properties** (interplay of thermal and quantum fluctuations, under what conditions universal scaling?)
1D Ising chain in transverse field

\[ H = - \sum_i J \, S_i^z S_{i+1}^z \]

2 ground states: \( \uparrow \uparrow \uparrow \uparrow \uparrow \uparrow \) or \( \downarrow \downarrow \downarrow \downarrow \downarrow \downarrow \)

- transverse field \( B \, S^x \sim - B \, (S^++S^-) \)
flips spins \( \uparrow \uparrow \uparrow \downarrow \downarrow \downarrow \downarrow \downarrow \uparrow \uparrow \)

\( \Rightarrow \) propagating solitons (Jordan Wigner fermions)

classical soliton (large-\( S \) limit)
Ising chain at criticality

- $\omega/T$ scaling expected, special “conformal” symmetry

- different universality class from Luttinger liquids (1D Heisenberg and XY AFM chains)

gapless linear (Dirac) spectrum

$\omega = c|k|$

for critical solitons
Experimental requirements

1) good 1D character to see solitons
2) low-exchange $J \sim 1$ meV to access critical field $BC \sim J/2 < 10$ T
3) strong uniaxial anisotropy (Ising character) but not perfect to still have transverse $g$-factor

- best Ising magnets are based on Co$^{2+}$ 3d$^7$

lowest Kramers doublet effective spin-1/2 Ising-like

Strong Crystal field + Spin Orbit

$\lambda \, L \cdot S$

2D Ising AF K2CoF4 \textit{(Birgeneau ‘73, Cowley ‘84)}
1D Ising AF CsCoCl3 \textit{(Goff ’95) also CsCoBr3 (Nagler)}

$J \sim 12$ meV $BC > 50$ T not accessible
Quasi-1D Ising ferromagnet CoNb$_2$O$_6$

zig-zag Co$^{2+}$ spin chain along c

Ferromagnetic superexchange
\sim 90^\circ \text{ bond Co-O-Co}
\sim 20K \sim 2\text{meV}

Ferromagnetic order along chain
Strong easy-axis (Ising) in \textit{ac} plane

Single crystal of CoNb$_2$O$_6$
(Oxford image furnace)

[Diagram of the crystal structure with Co$^{2+}$ ions and oxygen ions, showing the zig-zag chain and the ferromagnetic superexchange bonds.]
Magnetic excitations in 1D phase seen by neutron scattering

$T = 5 \, \text{K}, \ 1\text{D phase above } T_N$

- gapped continua characteristic of 2-soliton excitations

RC et al, Science 327, 177 (2010)
Magnetic excitations in zero field

- rich structure: continuum as characteristic of 2-soliton excitations
  + sharp modes (bound states)

RC et al, Science 327, 177 (2010)
Excitations have 1D character – no measurable dispersion \( \perp \) chains

\[ \parallel \text{chain direction} \]

\[ \perp \text{chain direction} \]
Zeeman ladder of bound states in 3D ordered phase

Continuum of free 2-soliton states

Bound states in confining potential

Soliton separation costs energy:

\[ V(x) = \lambda x \]

\[ \lambda \sim J' \langle S^z \rangle \]

Longitudinal mean-field: \(-hS^x\), \(\lambda = 2h\langle S^z \rangle\)
Zeeman ladder of bound states in 3D ordered phase

Continuum of free 2-soliton states

Bound states in confining potential

Soliton separation costs energy

$V(x) = \lambda \cdot x$

$\lambda \sim J' \langle S^z \rangle$

Longitudinal mean-field $-hS^z$, $\lambda = 2 \, h \langle S^z \rangle$
Zeeman ladder of bound states in 3D ordered phase

$T = 5 \text{ K}$

$T = 40 \text{ mK}$

Continuum of free 2-soliton states

Bound states in confining potential

Soliton separation costs energy

$V(x) = \lambda \cdot x$

$\lambda \sim J' \langle S^z \rangle$

Longitudinal mean-field $-hS^z$, $\lambda = 2 \ h \langle S^z \rangle$
**Soliton confinement**

*Mccoy&Wu* (’78)

**Schrödinger’s equation**

\[-\frac{\hbar^2}{\mu} \frac{d^2 \varphi}{dx^2} + \lambda |x| \varphi = (m - 2m_o) \varphi\]

**kinetic energy**    **string tension**

\[m_j = 2m_o + z_j \lambda^{2/3} \left( \frac{\hbar^2}{\mu} \right)^{1/3}\]

**Ai** function

\[\text{Ai}(-z_n) = 0\quad \text{Airy function}\]

\[z_n = 2.33, 4.08, 5.52, 6.78…\]

**Energy (meV)**

0.0 1.0 1.2 1.4 1.6

**Intensity (arb units)**

0.0 0.1 0.2 0.3

\[\hbar^2/(2\mu \tilde{c}^2) = 0.23(2) \text{ meV}\]

\[\lambda \tilde{c} = 0.033(1) \text{ meV.}\]
Phenomenological model of soliton gas

- work perturbatively around the Ising limit (spin clusters)

\[ J \uparrow\uparrow\downarrow\downarrow\downarrow\uparrow \uparrow \quad \text{2-soliton states} \]

\[ H = J \left| \uparrow\uparrow\downarrow\downarrow\downarrow\uparrow \uparrow \right> \quad \text{a 2 soliton state} \]

\[ -\alpha \left| \uparrow\uparrow\downarrow\downarrow\downarrow\downarrow\uparrow \right> + \ldots \quad \text{soliton hopping} \]

\[ -\beta \delta_{n,1} \left( \left| \uparrow\downarrow\uparrow\leftarrow \right> + \left| \uparrow\uparrow\uparrow\uparrow \right> \right) \quad \text{from XY term} \]

\[ -J_{xy} \left( S_i^x S_{i+1}^x + S_i^y S_{i+1}^y \right) \sim S_i^+ S_{i+1}^- + S_i^- S_{i+1}^+ \]

RC et al, Science 327, 177 (2010)
Phenomenological model of soliton gas describes full spectrum

\[ T = 40 \text{ mK} \]

**Gap:** \( J \sim 1.94 \text{ meV} \) from Ising \( zz \) exchange

**Bandwidth** \( \alpha = 0.12 \, J \) domain-wall hopping term

[ microscopic origin \( S^z S^x \ldots ? \)]

**Kinetic bound state:** transverse couplings for nn bond \( S^x S^x + S^y S^y \), \( J^\perp / J^z = 0.24 \)

and 2-nd neighbour AFM along chain \( J^{zz'} = -0.15 \, J^z \)

**Weak confinement term:** \( h_z \sim 0.02 \, J \) longitudinal field includes interchain mean-field numerical calculation agrees with exact analytic solution of effective Hamiltonian

Experiments in applied transverse field

Gap decreases with field

- field tunes quasiparticle dispersion

Field $\sim$ kinetic energy

$B_x S^x = (S^+ + S^-)/2$

Place crystal in metallic cage to prevent movement under high torque

$\begin{array}{l}
\text{CoNb}_2\text{O}_6 \\
torque
\end{array}$
Excitations as a function of transverse field

Two-soliton states

Ordered

Paramagnetic

Excitations change character above critical field

Magnetic 3D LRO

Bragg peak

Counts (10^3/sec)

Energy (meV)

Intensity (a.u.)
Excitations in transverse field

2-soliton continuum

RC et al, Science 327, 177 (2010)
Summary

- realized experimentally field-tuned quantum phase transition in quasi 1D Ising magnet CoNb$_2$O$_6$

- observed transmutation of quasiparticles at critical point
Conclusions

- neutron scattering is a very versatile probe of magnetic ordering and dynamics (\(\mu\text{eV}->\text{eV}\), magnetic field (15 T->25 T), low T (mK)
- quantitative: probe dispersions of excitations through well-understood matrix element (quantitative comparison with theories)
- sample size limited, large crystals needed (advanced crystal growth)
- new neutron sources (ISIS 2\(^{\text{nd}}\) target station, USA + JAPAN, ESS) will bring new opportunities: higher flux, higher resolution, wider coverage

50 detectors, 2D data set

40,000 detectors, 4D data set, 4 energies measured simultaneously,

- complementary to resonant x-ray diffraction and inelastic (RIXS) (samples 10’s of \(\mu\text{m}\), resolution >30 meV, T > few K – beam heating)
**Collaborators**

**La$_2$CuO$_4$**
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