High resolution backscattering instruments

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In this talk I present the principle of operation of indirect-geometry time-of-flight spectrometers such as IRIS at the ISIS spallation neutron source. The key features that make those types of spectrometers ideally suited for low-energy spectroscopy are: high energy resolution over a wide dynamic range, and simultaneous measurement over a large momentum transfer range provided by the wide angular detector coverage. To exemplify these features, I present results of single-crystal experiments of the spin-dynamics in the two-dimensional frustrated quantum magnet Cs_2CuCl_4 [1].

Strongly-correlated quantum systems often display novel collective behaviour that cannot be described in terms of the single-particle properties but rather is a consequence of correlations between infinite numbers of interacting particles. Such *emergent* quantum phenomena include spin-charge separation in 1D conductors or the fractional quantum Hall effect (FQHE) in the 2D electron gas in high magnetic fields. Elementary excitations often have unusual properties such as fractional quantum numbers and examples include the $1/3e$ vortices in the FQHE. Quantum magnets allow very quantitative studies of the nature of quantum states since neutrons can couple directly to the excited spin states. Here I present an investigation of a strongly-frustrated 2D quantum magnet, Cs_2CuCl_4, which has an exotic ground state with strong quantum fluctuations leading to large renormalizations of both static properties such as incommensurate ordering wavevectors and dynamic properties such as excitation energy scales. The elementary excitations are spinons, particles carrying *half* the spin of an integer spin flip, leading to extended continua in the dynamic correlations.

Cs_2CuCl_4 has spin-1/2 Cu ions arranged in magnetic layers where they form a frustrated triangular lattice as shown in Fig. 1. As opposed to un-frustrated systems such as the simple square lattice, well understood theoretically and studied experimentally in great detail [2], frustrated 2D quantum physics is theoretically very poorly understood and experimentally unexplored. The Hamiltonian in Cs_2CuCl_4 has not been realised in nature before and is expected to show exotic quantum behaviour because frustration tends to enhance rather than suppress quantum fluctuations. A comprehensive experimental study was made using high magnetic fields to manipulate the eigenstates and control the ground-state fluctuations, and use neutron scattering to probe the matrix elements of the transitions between eigenstates.

Due to the frustrated couplings the magnetic ordering that occurs at low temperatures is incommensurate along the chain direction b and the ordered spins rotate in helixes [3]. Fig. 2(c) shows a front view of a magnetic layer where ordered spins rotate mainly in the plane of the layer due to a small confining anisotropy. To determine the magnetic Hamiltonian measurements were made in the fully-polarised state at high fields [1]. This states is an exact eigenstate of the Hamiltonian with no fluctuations where excitations are one-spin flip magnons with dispersion equal to the Fourier transform of the couplings, therefore all exchanges could be determined *exactly*. Measurements at 12 T $\|$ a [1] reveal a strongly coupled 2D quantum magnet with $J'=0.33J$ and small anisotropy terms of order 5% J. Zero-field measurements observe a large quantum renormalization of the incommensurate ordering wavector $\varepsilon_0=0.56\ \varepsilon_C$, where ε_C is the classical incommensuration observed directly in the fully-polarized state where fluctuations are quenched out.

High-resolution studies of the excitation lineshapes were made using the IRIS spectrometer at the ISIS spallation neutron source, see Fig. 3. The indirect geometry combined with a long incident flight path and near back-scattering at the analysers give a very high energy resolution ~15μeV. The dynamic range that can be adjusted by phasing the two disk choppers in the incoming beam. The large angular bank allows coverage over a large range of momentum transfers such that the dynamic response can be measured in several Brillouin zones simultaneously. Typical measurements of the excitation lineshapes are shown in Fig. 4.(a) where the gray patch shows the phase space covered by a group of detectors at a given scattering angle 2θ projected onto the reciprocal space of the sample and the middle panel gives the intensity measured along this line. The absence of single-particle peaks and presence of strong excitation continua is the characteristic feature of fractionalization. Resolution effects are minimal as illustrated by the narrow instrumental resolution shown in the figure. Since neutrons have spin-1/2, in a scattering process they can transfer a total spin ΔS=0 or 1 to the systems and the absence of single-particle poles and the presence of strong excitation continua show that the elementary excitations carry fractional spin quantum number. By analogy to the pure 1D case where the dynamic correlations also show broad continua, we identify the quasiparticles in Cs_2CuCl_4 with spin-1/2 spinons.

The measured lineshapes can be used to construct a map of the magnetic scattering throughout the zone and Fig. 5 shows the dispersion relations of the lower and upper boundaries of the scattering continua (dotted areas) along the *b*-axis. The dispersion of the main peak in the scattering lineshape is indicated by the solid points and is incommensurate, with minima at the incommensurate positions of the magnetic Bragg peaks, and asymmetric with 2 non-equivalent zone boundary energies of 0.55 and 0.7 meV. These are features of the frustrated couplings and can be explained by a spin-wave model for the (J,J') Hamiltonian. The extracted effective exchange couplings show that the energy scale of the magnetic excitations is renormalized due to quantum fluctuations by a factor R=1.65, denoting strong quantum fluctuations in the ground state.

The existence of spinons in two-dimensional systems has been an outstanding problem in strongly-correlated magnetism. In 1973 Anderson proposed that a 2D fractional quantum spin liquid would take the form of a resonating valence bond (RVB) state [4]. To first order the ground state consists of singlet spin pairings [see Fig. 6(a)] and higher-order corrections allow these bonds to fluctuate or "resonate" between different neighbours. Breaking up a bond in a neutron scattering process creates two free spin-1/2 "spinons" that can propagate away independently through a rearrangement of the intervening bonds [see Fig. 6(b)].

In conclusion, indirect-geometry time-of-flight spectrometers are ideally suited for low-energy single-crystal spectroscopy studies. High resolution over a wide dynamic range, large momentum space coverage and low background (cooled analyzers) allow quantitative measurements of the excitation lineshapes and dispersion relations. Enabling high magnetic field studies and polarization analysis would give unprecedented quantitative information about the nature of eigenstates in quantum magnets and other strongly-correlated systems.

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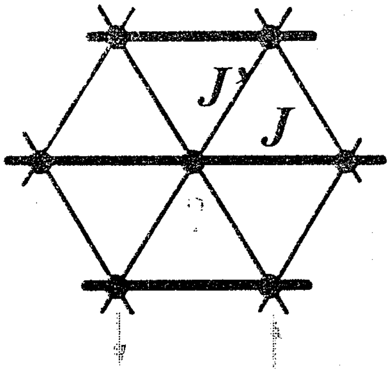
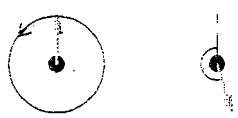
Introduction



2D frustrated quantum magnet Cs_2CuCl_4

J'/J=0.33

- frustrated 2D quantum physics is experimentally unexplored and theoretically very challenging



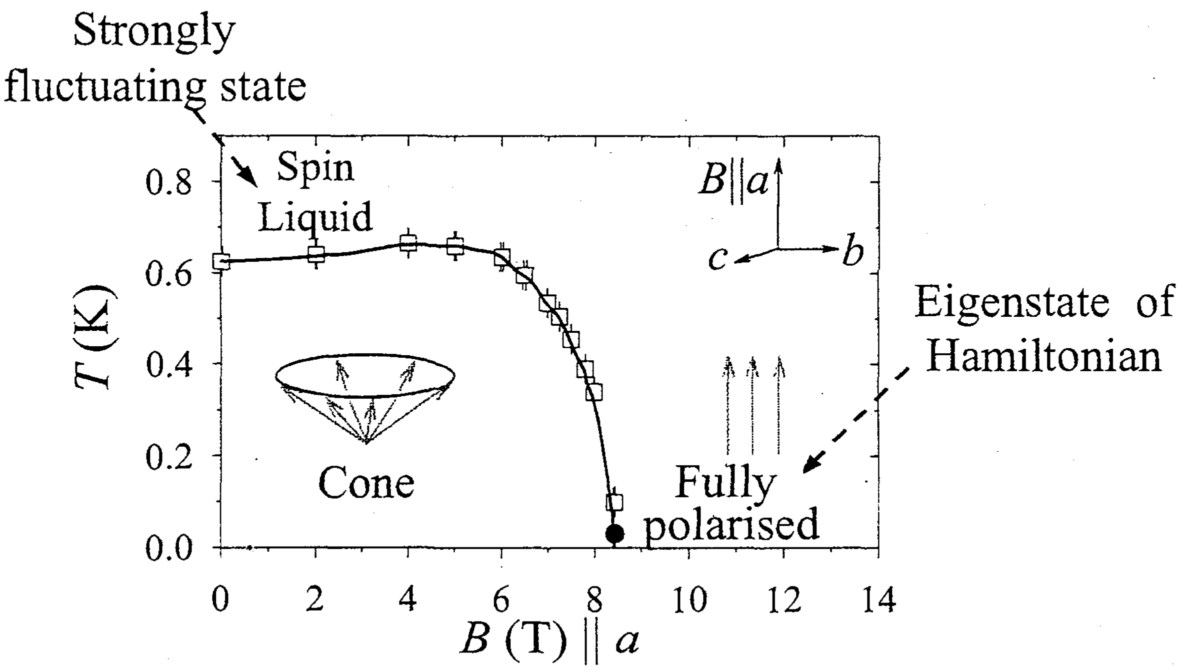
Incommensurate, non-collinear spin correlations

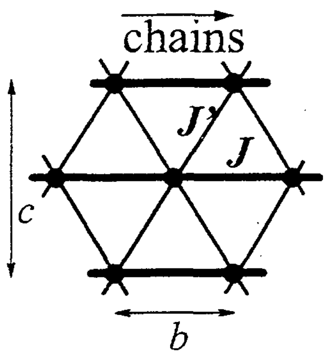
- Hamiltonian not been realised in nature before and expected to show exotic quantum behaviour

<u>Experimental approach:</u>

- use magnetic fields to manipulate the eigenstates and control the quantum fluctuations
- use neutron scattering to probe the matrix elements

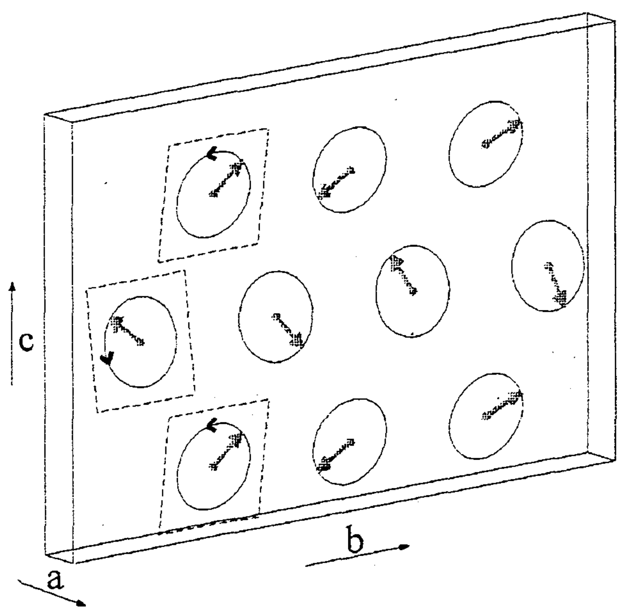
Magnetic phase diagram ($B||a$)





Cs_2CuCl_4

Spins rotate in cycloids in zero field.

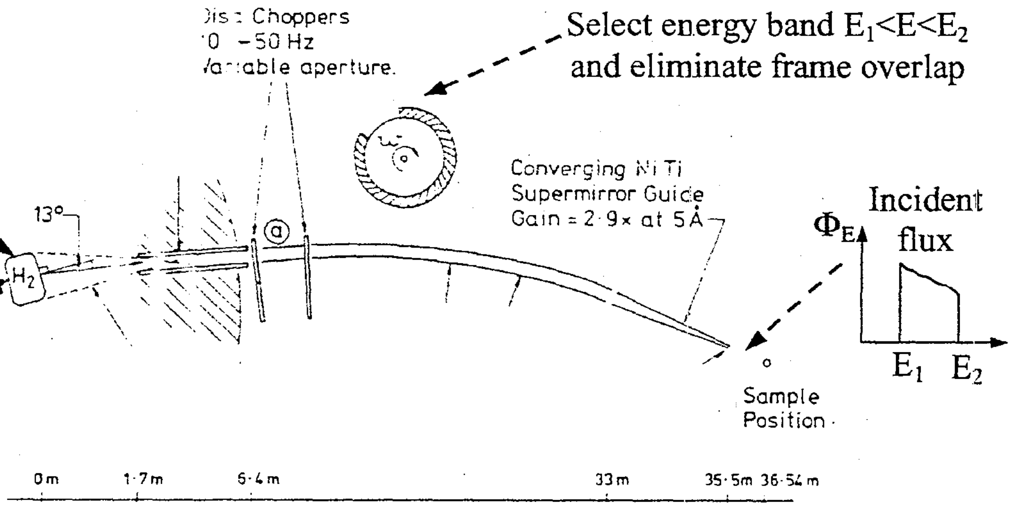


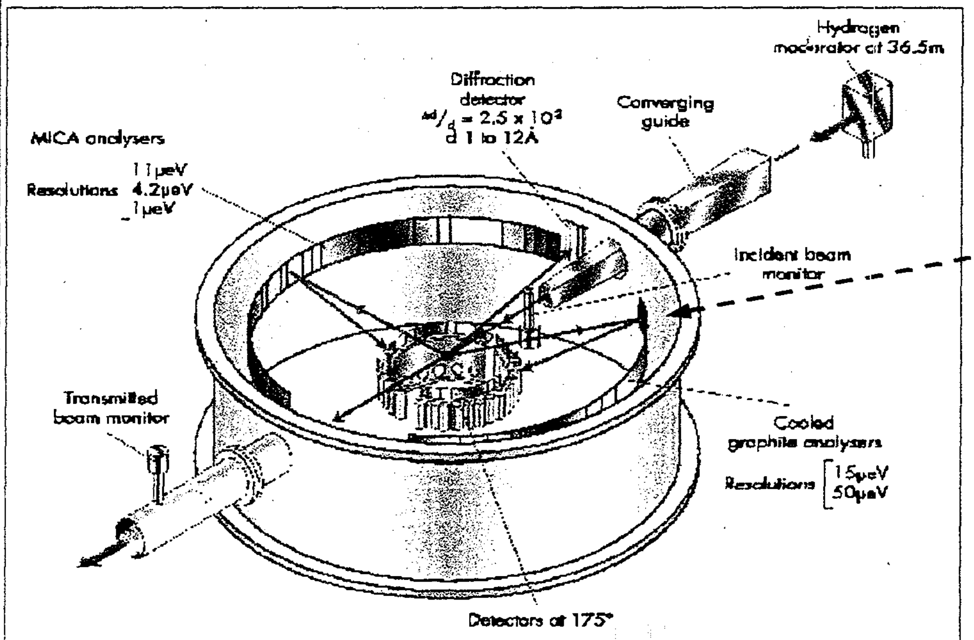
The IRIS spectrometer at ISIS, UK

Select energy band $E_1<E<E_2$ and eliminate frame overlap

Liquid H_2 moderator (25 K)

Target (neutron pulses produced every 20 ms)





Bragg scattering at analyser select fixed E_f

$d \sin\theta = \lambda_f$

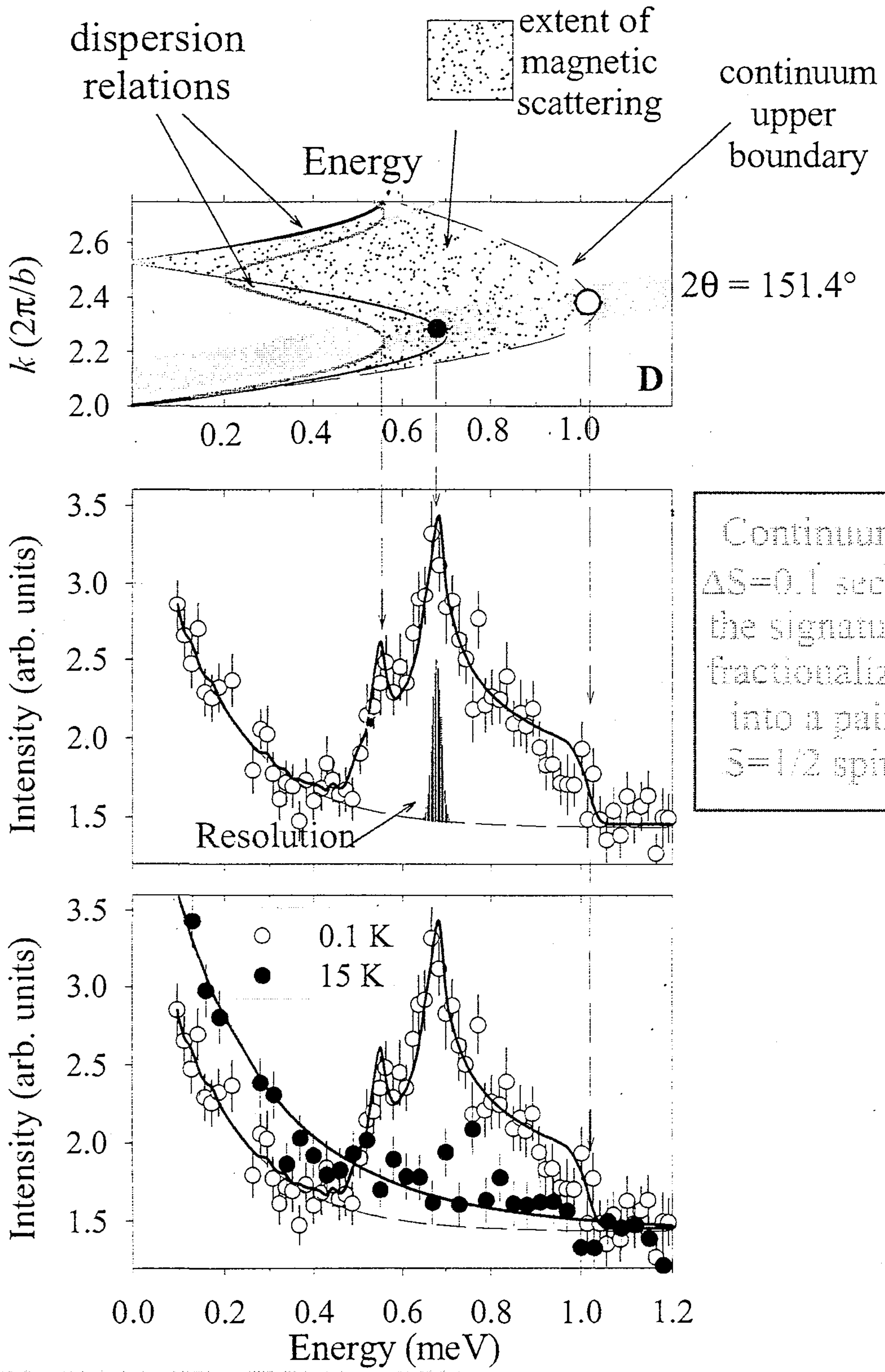
Energy resolution : $\Delta E/E \sim 2\,[\,\Delta t_m/t\,] * [\,\Delta d/d\,] * [\cot\theta \cdot d\theta]$

Long flight path (t large)

Near backscattering $2\theta = 175°$

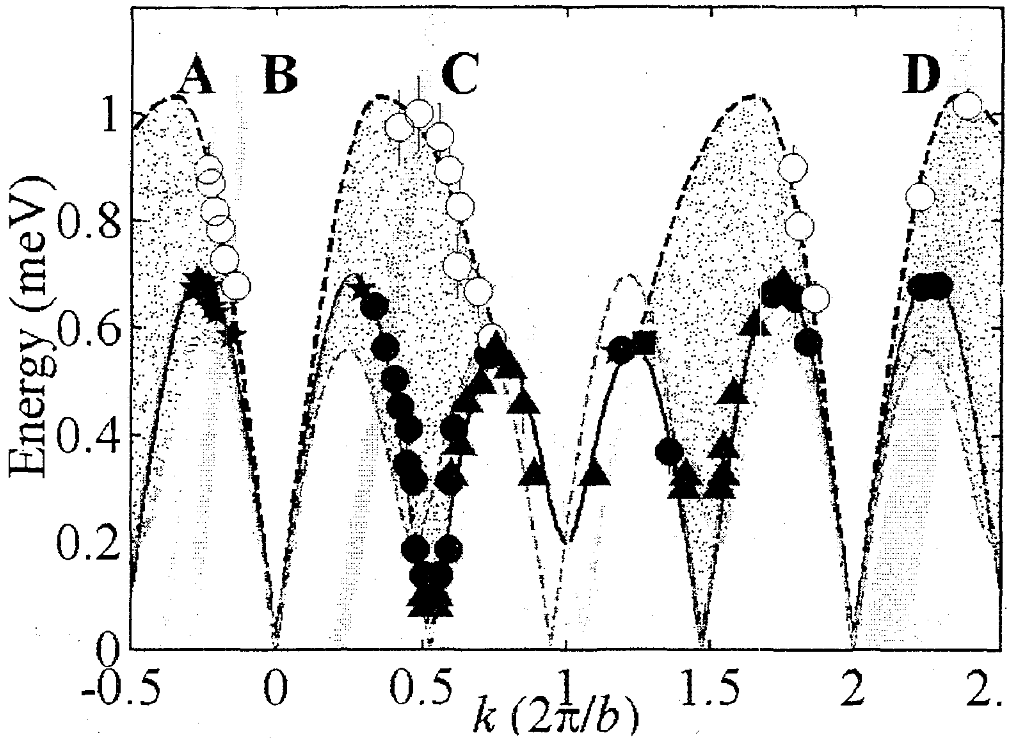
High resolution $\Delta E \sim 15$ μeV
Wide dynamic range -0.2 to 1.6 meV (variable)
Large angular coverage $25° < 2\theta < 158°$

Excitations in zero field

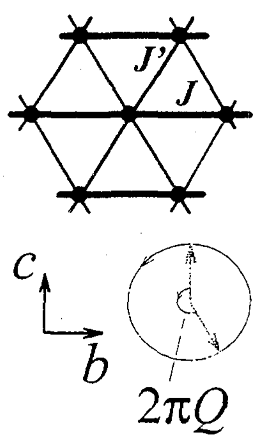


Continuum in $\Delta S=0,1$ sector is the signature of fractionalization into a pair of $S=1/2$ spinons

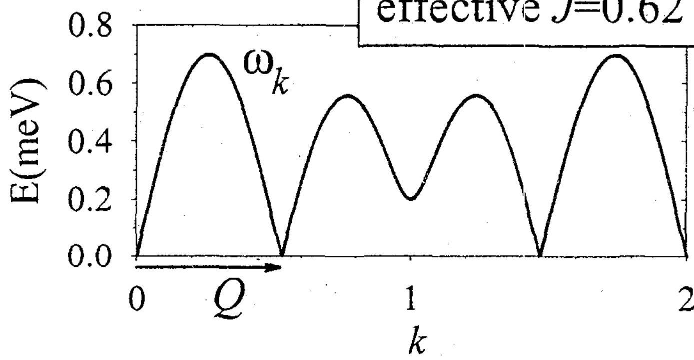
Dispersion relation (B=0)



Spin-wave model



effective $\tilde{J}$=0.62 meV



J=0.37 meV

ω_k polarized out-of-plane, ω_{k+Q} polarized in-plane

Quantum renormalization of excitation energy

$R=\tilde{J}/J=1.65$

(1D S=1/2 HAFC $R=\pi/2$)

Resonating Valence Bond Spin Liquid