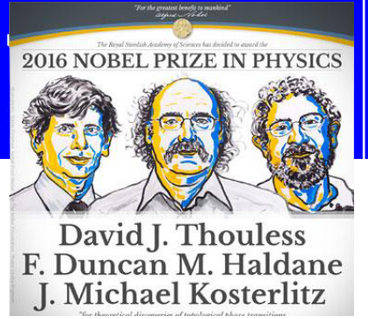
ILL news
DECEMBER 2016
special issue

HALDANE RECEIVES NOBEL PRIZE IN PHYSICS FOR HIS WORK IN THE ILL THEORY GROUP

NEUTRONS FOR SOCIETY



Early ILL neutron scattering evidence for a new quantum state

The quantum behaviour of interacting many-particle assemblies leads to surprising and counter-intuitive new phenomena, superconductivity and superfluidity are well-known examples.

Duncan Haldane discovered a new quantum mechanical phenomenon as post-doc at the ILL [1]: one-dimensional antiferromagnets ("spin chains") would behave entirely different depending on whether the individual spins are integers (1,2,3,...) or half-integers (1/2, 3/2, ...).

This fundamental distinction is inherently related to the fact that one turn brings an integer spin back to its initial state, while a half-integer spin needs two turns. This appears quite esoteric, however, Haldane made precise predictions that could directly be tested by neutron scattering (see cartoon): integer spin chains should display only short-ranged pair correlations and a triplet excitation above a large energy gap, while half-integer chains should show infinitely-far reaching correlations (leading to long-range order in presence of even tiny interchain coupling), and gapless excitation spectra. Hence a spin-1/2 antiferromagnetic chain seems closer to the "classical" antiferromagnetic Neel long-range order than the spin-1 chain. Since quantum effects scale with 1/S, nobody had expected anything else than an increasing tendency towards long-range order with increasing spin value. While theorists at the ILL [2] and round the world were working hard to elaborate this puzzle, the new idea was immediately tested experimentally, and early ILL experiments provided "proof" prior to a full theoretical understanding [3].

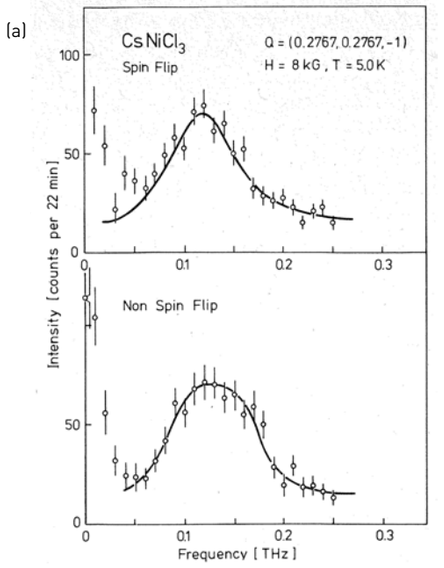


Fig. 1 $CsNiCl_3$
(a) Isotropic gapped excitation [5], IN12.
(b) Zeeman splitting of the Haldane triplet [6]. IN20.

In real materials, excitation gaps arise for a variety of reasons, amongst which anisotropy and dimerisation were well-known at the time. The first evidence for the "Haldane gap" was found in the quasi-one dimensional spin-1 compound $CsNiCl_3$ at a temperature where the small interchain interaction is overcome by thermal fluctuations [4]. The argument was based on the absence of quasi-elastic scattering and the discrepancy of anisotropies derived from perturbation theory compared to those from linear spin-wave theory. Polarised IN12 experiments (fig.1a) provided the first direct proof of the isotropic triplet character of the gapped excitation in $CsNiCl_3$ [5] and excluded an anisotropy origin. Polarised IN12 and IN20 data (fig.1b) proved Zeeman splitting of the triplet [6] and triplet character all along the one-dimensional dispersion [7,8]. In parallel, a new spin-1 compound with much smaller interchain interaction was studied, NENP ($Ni(C_2H_8N_2)_2NO_2(ClO_4)$), which in contrast to $CsNiCl_3$ did not order at any temperature. Nevertheless, initially, the origin of the excitation gap was not clear due to a large anisotropy, and dimerisation could not be excluded either [9]. The precise ratio between anisotropy and exchange was established by additional experiments on IN12,IN8,IN20 and ruled out a trivial anisotropy gap [8-11].

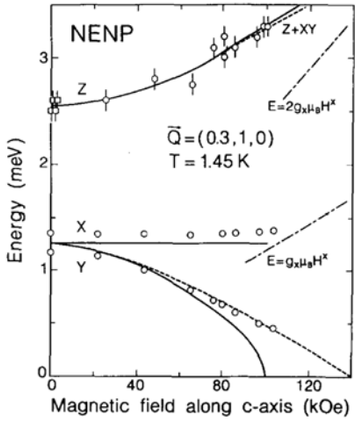


Fig.2 NENP ($Ni(C_2H_8N_2)_2NO_2(ClO_4)$)
Zeeman effect of the anisotropy-split triplet DN1, IN12,IN8,IN20 [11].

Fig.2 shows the Zeeman splitting of the three gapped excitations [11]. Soon French chemists synthesised two spin chain compounds of identical crystallographic structure, $AgVP_2S_6$ and $AgCrP_2S_6$, with spin 1 (V) and spin 3/2 (Cr), respectively. Experiments on D1B and IN4 showed long-range order and gapless excitations for the spin 3/2 compound (fig.3a), and absence of long-range order and an excitation gap for the isostructural spin-1 compound (fig.3b). This was the first direct evidence for the distinction between integer and half-integer spin chains [12]. Y_2BaNiO_5 is the spin-1 chain with

the smallest anisotropy and the best one-dimensionality known today. It was discovered and characterised by Spanish scientists on D2B [13].
Numerous neutron experiments at the ILL and elsewhere dealt with further details and related phenomena.

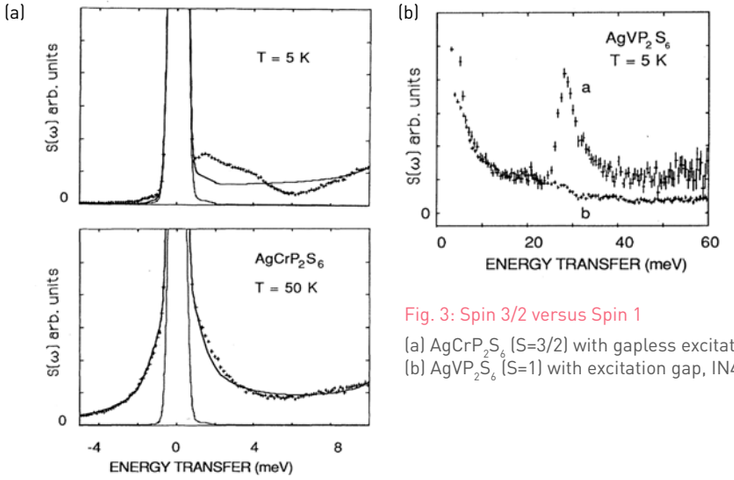


Fig. 3: Spin 3/2 versus Spin 1
(a) $AgCrP_2S_6$ (S=3/2) with gapless excitations, IN4.
(b) $AgVP_2S_6$ (S=1) with excitation gap, IN4 [11].

Duncan Haldane pointed out [1] that the one-dimensional quantum spin-1 antiferromagnet at zero temperature can be related to the classical two-dimensional planar ferromagnet at finite temperature. According to Kosterlitz and Thouless, the latter has vortex-like topological excitations ("skyrmions") that condense into the ground-state upon lowering the temperature. The quantum spin-1 chain's ground state is characterised by a topological non-local order, which is analogous to the topological order responsible for the fractional quantum Hall effect [14] (cartoon). Fig. 4 and 5 show spectra of isotropic one-dimensional antiferromagnets with spin 1/2 and 1 respectively, taken on modern ILL instruments (IN5,IN4).

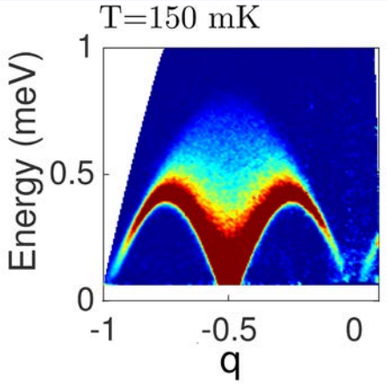


Fig. 4 Spin 1/2: gapless spinon continuum $CuSO_4.5D_2O$ measured on IN5

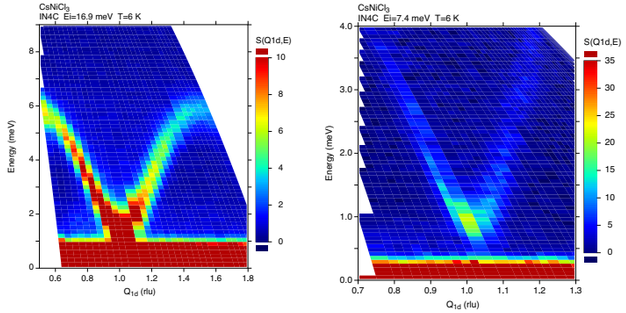


Fig. 5 Spin 1: : sharp and gapped excitation spectrum $CsNiCl_3$ measured on IN4
left: sharp dispersion, right (with higher resolution): gap.

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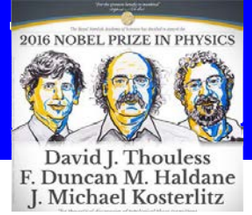
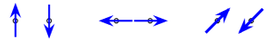


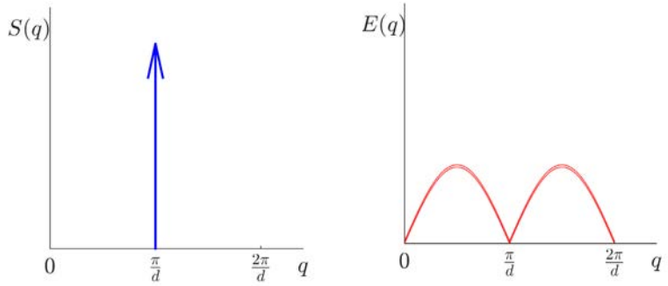
Haldane for beginners (isotropic case):

Consider isotropic magnetic interactions between nearest-neighbour spins that favour antiparallel alignment independent of the spin direction in space:



At T=0, an isotropic classical (i.e. S→ ∞) one-dimensional antiferromagnet is in the Neel state:

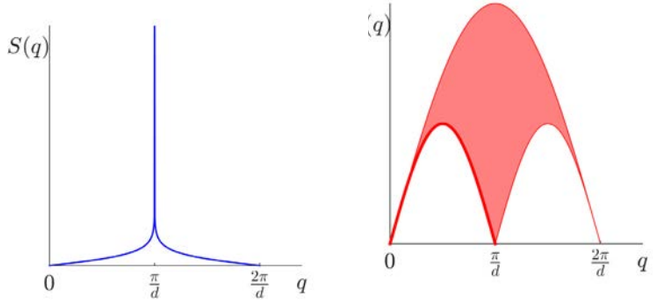
This pattern leads to sharp Bragg peaks in diffraction.



The excitations are doubly degenerate spin waves and correspond to small deviations from the ordering direction. Their energy-momentum relation (dispersion) has no energy gap.

For quantum spins 1/2 the Neel state is NOT the ground state, but a snapshot would still show infinitely large regions that look like the classical Neel state.

There are no Bragg peaks, but the peak intensity still diverges in a power-law, corresponding to an infinite correlation length like at a critical point.

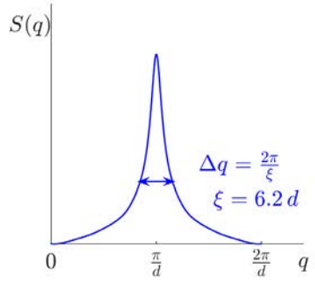


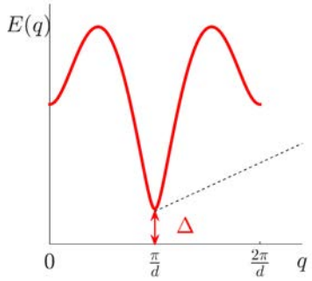
The excitations are pairs of topological (domain wall like) spinons with a gapless dispersion.

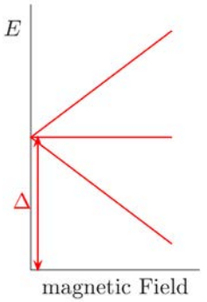
Even a tiny interchain coupling leads to long-range order in real spin 1/2 materials.

Haldane predicted that integer spin (1,2,3,...) antiferromagnetic chains appear even less ordered,

With broad non-diverging peaks in diffraction.





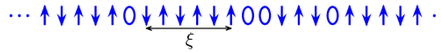


The lowest excitation has a finite energy ("Haldane gap"). It is sharp in energy and triply degenerate.

Half-integer (1/2, 3/2, ...) antiferromagnetic spin chains should resemble the gapless spin 1/2 case.

For spin 1, the Haldane ground state singlet is characterised by a topological non-local (string) order. This string order can be expressed by the recipe: spin up (Sz=1) needs to be followed by spin down (Sz=-1) or any number of zeros (Sz=0), and spin down by spin up or any number of zeros. Antiferromagnetic correlations remain short.

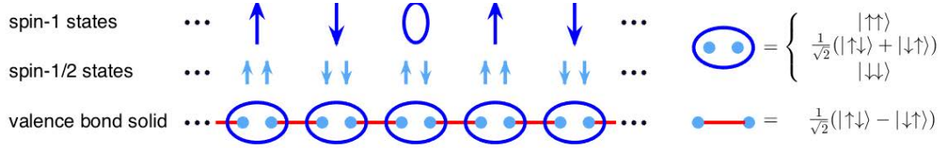
Ground state



The gapped "Haldane triplet" excitation ruptures the string order in a topological (domain-wall like) manner.

Triplet excitation

An equivalent picture for the string order is achieved by decomposing each spin 1 into two spins 1/2. On each site the spins 1/2 are paired symmetrically to a spin 1, between neighbouring sites they form a singlet pair state (antisymmetric pairing, "valence bond"). This "valence bond solid" reflects best the unbroken translation symmetry of the Haldane ground state singlet.



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