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

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 CsNiCl₃ 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 CsNiCl₃ [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(C₂H₈N₂)₂NO₂(ClO₄)), which in contrast to CsNiCl₃ 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 shows the Zeeman splitting of the three gapped excitations [11]. Soon French chemists synthesised two spin chain compounds of identical crystallographic structure, AgVP₂S₆ and AgCrP₂S₆ 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.3b), and absence of long-range order and an excitation gap for the isostructural spin-1 compound (fig.3a). This was the first direct evidence for the distinction between integer and half-integer spin chains [12]. Y₂BaNiO₅ 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.

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. 3: Spin 3/2 versus Spin 1](image)

(a) AgCrP2S6 (S=3/2) with gapless excitations, IN4.
(b) AgVP2S6 (S=1) with excitation gap, IN4 [11].

![Fig. 4: Spin 1/2: gapless spinon continuum CuSO4.5D2O measured on IN5](image)

![Fig. 5: Spin 1: sharp and gapped excitation spectrum CsNiCl3 measured on IN4](image)

(left) sharp dispersion, right (with higher resolution): gap.

References:

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:

\[ \ldots \quad \uparrow \quad \downarrow \quad \uparrow \quad \downarrow \quad \ldots \]

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.

\[ \ldots \quad \uparrow \uparrow \quad \downarrow \downarrow \quad \ldots \]

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.

Haldane predicted that integer spin (1, 2, 3, ...) antiferromagnetic chains appear even less ordered, whereas half-integer (1/2, 3/2, ...) antiferromagnetic spin chains should resemble the gapless spin 1/2 case.

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.

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.

Even a tiny interchain coupling leads to long-range order in real spin 1/2 materials.
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Our very best wishes for the year ahead from all at the ILL

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