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# Interacting quantum spin chains

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**Abstract.** A brief review of recent advances in neutron scattering studies of low-dimensional quantum magnets is followed by a particular example. The separation of single-particle and continuum states in the weakly coupled $S = 1/2$ chains system $BaCu_2Si_2O_7$ is described in some detail.

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For the last two decades, low-dimensional quantum magnets have been the subject if intensive neutron scattering studies. One of the main reasons for this steady interest is that low-dimensional systems are *simple* models of magnetism that demonstrate a broad spectrum of *complex* quantum-mechanical phenomena. In many cases quantum magnets are desribed by simple Hamiltonians with few parameters. Theoretical and numerical studies of these models can be directly compared to experiment at the quantitative level, often yielding remarkable agreement, and provide guidance in the data analysis. Neutron scattering techniques are particularly well suited for studying real low-dimensional magnets. Indeed, they provide *direct* measurements of the spin correlation function $S(\omega)$, that carries significant physical information and is the ultimate result of most theoretical calculations. Moreover, in most known low-dimensional magnets the energy and length scales of magnetic interactions perfectly match those probed by thermal or cold neutrons. It will not be an overstatement to say that the development of the entire field of low-dimensional magnetism has been driven by neutron experiments more than by any other experimental technique.

Two decades of research and huge amounts of beam time yielded a fairly complete understanding of the most basic one-dimensional models. To mention only a few milestones, we have to recall the study of local excitations in dimer systems [1], the discovery of the famous Haldane gap [2] and the observation of continuum excitations in $S = 1/2$ Heisenberg antiferromagnets (AFs) [3, 4]. A number of remarkable discoveries were made only recently. Among these are studies of multi-magnon excitations [5], observation of field-induced incommensurability in $S = 1/2$ systems [6], the spin-Peierls compound $CuGeO_3$ [7], continuum states [8] and field-induced ordering [9] in Haldane-gap antiferromagnets, and the effect of staggered fields on quantum spin chains [10]. These new studies were enabled by the discovery of new model materials, the development of new experimental techniques and the perfection of data analysis procedures.

Today, the general trend in low-dimensional magnetism is to capitalize on the accumulated knowledge of the basics and move on to more complex problems. Among the new and rapidly progressing directions of research are effects of randomness and doping in quantum spin chains [11–13], the interplay between charge and spin degrees of freedom [14], new physics in highly frustrated quantum antiferromagnets [15], and the crossover regime from "quantum" to "classical" magnetism. In this paper we cover as many of these new studies as possible. To keep it readable, however, below we shall concentrate on just one example, namely the dimensional crossover in weakly interacting $S = 1/2$ Heisenberg spin chains.

At the heart of the matter is a very old controversy. As far back as 1931 H. Bethe *exactly* solved the ground state of the one-dimensional (1D) $S = 1/2$ quantum Heisenberg antiferromagnet [16]. The main result was that even at $T = 0$ there is no long-range order in the system, and no Bragg peaks should be visible in a neutron diffraction experiment. A year later, L. Néel proposed the famous two-sublattice model of antiferromagnetism [17], characterized by staggered long-range magnetic order that produces new magnetic Bragg peaks in the diffraction pattern. In 1933 L. Landau published yet another paper on the subject, stressing the fact that the two-sublattice model is not even an eigenstate of the Heisenberg Hamiltonian, and therefore cannot *possibly* be the ground state [18]. Now we of course know that for a vast majority of 2D and 3D materials, the ground state does indeed look remarkably *like* the Neel state. Landau's arguments are *also* correct, and quantum fluctuations are relevant. In two and three dimensions they usually result in minor corrections. The

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lower the effective dimensionality, the more these fluctuations are important, and in the purely 1D case they are capable of destroying long-range order altogether. It is now well understood that *weakly coupled* $S = 1/2$ Heisenberg chains are *weakly ordered*: the Neel temperature $T_N$ scales roughly as the strength of inter-chain coupling $J'$, while the sublattice saturation moment at $T \to 0$ behaves as $J'/J$, $J$ being the in-chain exchange constant. Both quantities vanish as $J' \to 0$. It is important to note that long-range ordering occurs for arbitrary small $J'$. For example, correlated glassy freezing with an ordered moment of only 0.03 $\mu_B$ have recent been detected in the *extremely* one-dimensional material $SrCuO_2$ with $J'/J \approx 7 \times 10^{-4}$ [19].

The most interseting question is what happens to the excitation spectrum of a single $S = 1/2$ antiferromagnetic quantum spin chain when inter-chain coupling is "switched on". Let us first consider the extreme cases. In the 3D limit, when $J' \approx J$ we are dealing with a ground state that is very similar to the Neel state. The excitation spectrum is then dominated by single-particle states that correspond to a *presession* of the ordered moment around its equilibrium direction. These particles, known as *spin waves*, carry a total spin of unity and and a spin projection $S_z = \pm 1$ onto the direction of staggered moment. In the early days it was believed that in the other limiting case of a purely 1D AF the excitation spectrum is described by a similar single-particle picture, albeit with strongly renormalized spin-wave velocity and bandwidth [20]. It was later realized that spin dynamics in the 1D case is, in fact, *qualitatively* different. Since long-range order is absent, so are the precession modes. The spectrum contains *no* single-particle excitations and is instead a continuum of states [21–24]. An experimental confirmation of this phenomenon was obtained in elegant neutron scattering experiments on $KCuF_3$ [3] and copper benzoate [4]. Modern theories describe these continuum states as composed of pairs of exotic $S = 1/2$ excitations called *spinons*. Unlike magnons, which are bosons and can be directly observed in an inelastic neutron experiment, spinons carry $S = 1/2$, and are created or destroyed only in pairs, much like domain walls in an Ising magnet. The two-spinon continuum is three-fold degenerate with pairs of spinons having a total spin of unity and projections on any given axis $S_z = 0, \pm 1$. Note that while there are only two polarizations for spin waves, spinon pairs come in three different polarization flavors.

If the spin dynamics in the two limiting cases is *qualitatively* different, what happens in quasi-1D systems with $0 < J' \ll J$? The presence of long-range order should produce order-parameter excitations, i.e., spin waves. But how exactly are these single-particle states spawned from the continuum of inelastic scattering that dominates in the 1D system model? Theoretically, the problem is not a simple one: since isolated chains are *quantum-critical*, there is no "small" energy scale for inter-chain coupling. The latter, no matter how small, cannot be treated through any kind of infinitesimal expansion. Nevertheless, a very simple and accurate physical picture is provided by the chain mean field (MF) theory [25] and the random phase approximation [26, 27], when these approaches are combined with exact results for isolated chains in external staggered fields. In the ordered state each spin chain is subject to an effective staggered exchange field generated by neighboring chains. A staggered field $H_\pi$ induces a liner attractive potiential between spinons. As a result, the lowest-energy excitations are spinon *bound states*, often referred to as "magnons" [26, 27]. This is illustrated in Fig. 1b. The square root singularity on the lower bound of the two-spinon continuum in the isolated chains [Fig. 1a] "separates" and becomes a sharp magnon which is a $\delta$ function in energy at any given wave vector [Fig. 1b, solid line]. The magnons acquire a *gap* $\Delta$ (also referred to as *mass*) that scales as $H_\pi^{2/3}$. Since there are three possible spin states for a pair of spinons, there are *three* magnon branches. Two of these are polarized perpendicular to $H_\pi$ and the induced staggered moment, and

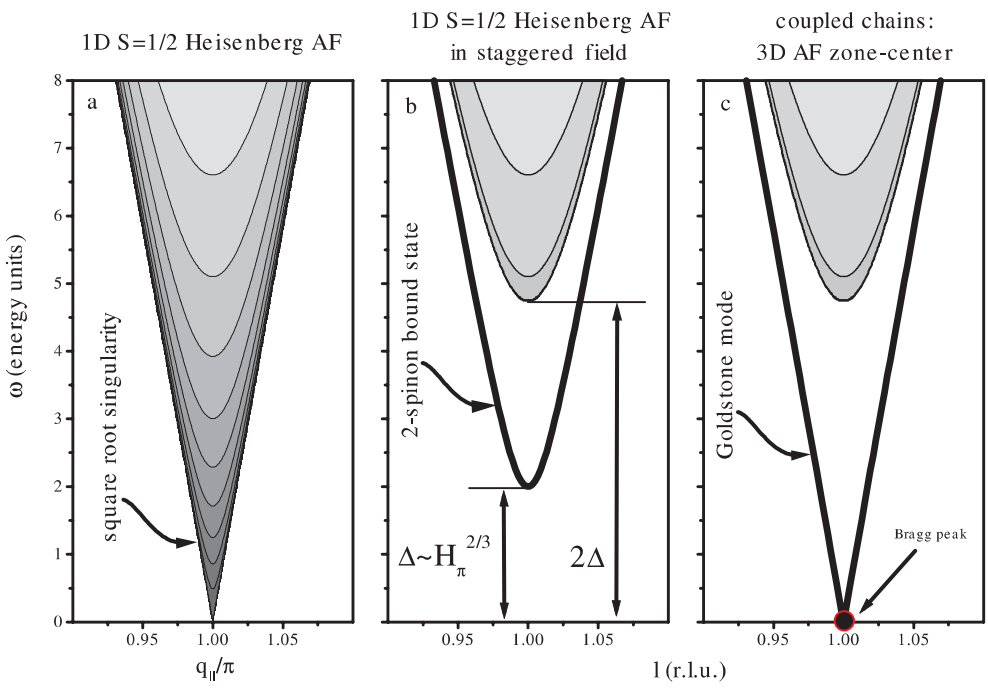


**Fig. 1a–c.** Transverse dynamic structure factor of the 1D $S = 1/2$ Heisenberg AF. (**a**) Contains only continuum excitations with a singularity on the lower bound. An external staggered field (**b**) induces a gap $\Delta$ in the spectrum. The singularity separates from the lower bound of the continuum giving birth to single-particle excitations. This picture is also observed in coupled chains at the transverse zone-boundary. In the latter case the single-particle states take the role of Goldstone modes (spin waves) and their energy goes to zero at the 3D AF zone center (**c**)

correspond to conventional precession modes (spin waves). Including inter-cahin interactions within the random phase approximation (RPA) gives these excitations a dispersion perpendicular to the chains. Their energy goes to zero at the 3D zone center, i.e., at the location of magnetic Bragg peaks in the ordered system [Fig. 1c, solid line]. The gap $\Delta$ can still be observed at the transverse zone boundary, where the behavior of an isolated chain in a staggered field is exactly recovered [Fig. 1b]. What remains of the two-spinon continuum in the 1D system is now seen as a *three-magnon*, rather than a *two-spinon* continuum. Indeed, the attractive potential between spinons is a *confining* one, and two spinons are permanently bound into magnons, just like two quarks can be confined in a meson. The continuum therefore has a gap of $2\Delta$, i.e., twice the characteristic magnon gap.

An experimental observation of such rich and unique behavior, the *separation* of single-particle and continuum states, is a formidable challenge to neutron scattering. On the one hand, a strongly 1D system with $J' \gg J$ is desirable to maximize the fraction of the spectral weight contained in the continuum, a feature notoriously difficult to observe. On the other hand, $J'$ should be large enough to yield a measurable gap $\Delta$ (preferably, a few meV). Finally, $J$ should be *small* enough to allow measurements with a wave vector resolution better than $\Delta/v$, where $v = \pi/2J$ is the spin-wave velocity. The two latter conditions are absolutely essential to resolving the magnons at energy $\Delta$ from the lower bound of the continuum at $2\Delta$. The first model system that met these conflicting requirements was $KCuF_3$, a material with $J = 17.5$ meV, $T_N = 39$ and a saturation moment of $m_0 \approx 0.5\ \mu_B$. In this compound the spin waves and continuum excitations could be observed simultaneously [28].

Below we shall make the experimental case for separation of single-particle and continuum states using another model quasi-1D material, namely $BaCu_2Si_2O_7$. In this compound $J = 24$ meV, $T_N = 9$ K and $m_0 = 0.15\ \mu_B$ [29, 30], i.e., $BaCu_2Si_2O_7$ is more one-dimensional than $KCuF_3$ is. The $S = 1/2$ AF chains run along the $c$ axis of the orthorhombic crystal structure. The 1D AF zone center $q_\parallel = \pi$ is the $(h, k, 1)$ reciprocal-space plane, and the magnetic Bragg peak, characteristic of 3D long-range ordering is located at (0,1,1). Despite the small saturation moment in $BaCu_2Si_2O_7$, its low-energy excitation spectrum (up to about 5 meV energy transfer) is dominated entirely by sharp single-particle spin-wave-like excitations [31, 32]. Very high resolution measurements performed using the IN14 cold-neutron spectrometer at ILL failed to detect any intrinsic excitation widths. Fig. 2 shows a series of constant-$q$ scans that measure the dispersion of these modes at the 1D AF zone-center in the direction perpendicular to the chain axis. The solid lines in Fig. 2 are a *global* fit to the data based on a single-mode cross section for a classical antiferromagnetic spin wave, convoluted with the spectrometer resolution function [32]. Measurements of the spin-wave dispersion along different reciprocal-space directions led to a fairly complete picture of inter-chain interaction [30]. The effective MF inter-chain coupling constant was found to be $J' = 0.4$ meV. The "magic point" where inter-chain interactions cancel out at the RPA level is located at $(0.5, 0.5, 1)$. The energy of the spin wave at this wave vector is to be interpreted as the gap $\Delta$ induced in each individual chain by their interactions with neighboring chains. Experimentally,

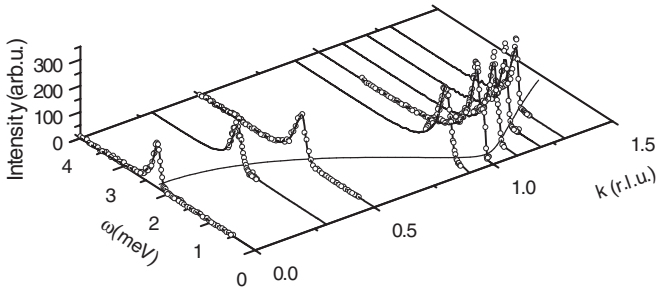


**Fig. 2.** A series of constant-$Q$ scans measured in $BaCu_2Si_2O_7$ at $T = 1.5$ K for different momentum transfers perpendicular to the chain axis. *Lines* represent a semi-global fit to the data as described in the text. The *solid lines* in the basal plane show the spin-wave dispersion relation in this reciprocal-space direction. The data are from [31]

for $BaCu_2Si_2O_7$, $\Delta = 2.5$ meV. The observed low-energy spectrum is totally consistent with theoretical predictions: at energies below $2\Delta$ transverse spin fluctuations in a weakly coupled chains system behaves exactly as those in a *classical* antiferromagnet.

The quantum-mechanical nature of the spin chains in $BaCu_2Si_2O_7$ becomes apparent on shorter time scales (larger energy transfers). Figure 3 shows a series of constant-energy scans accross the 1D AF zone center. At $\hbar\omega = 3$ meV, using the highest-resolution setup [Fig. 3a], one clerly sees two well-resolved peaks that represent the low-energy single-particle excitations. A fit of the classical spin-wave cross section to the data is shown by the shaded area. The two spin-wave peaks can not be resolved at $\hbar\omega = 3$ meV using a setup with coarser resolution [Fig. 3b]. However, at higher energies, [Fig. 3c–f] even the coarse-resolution configuration should have been capable of resolving two separate peaks if the single-particle picture still held (shaded areas). In contrast, the measured scans do not contain two separate peaks, but instead show a single broad feature. Moreover, the spin waves, for which intensity scales as $1/\omega$, are expected to account for only a very small fraction of the total spectral weight at high energy transfers [Fig. 3e,f]. The remaining scattering is to be attributed to the excitation continuum that sets in at about 5 meV energy transfer and becomes progressively more dominant at high energies. The bulk of the data collected in different experimental configurations was analyzed in a global fit using a cross section that contained both a single-mode and a continuum part. The cross section for the continuum was chosen to match the one calculated analytically within the Sine-Gordon model [27]. The continuum was assumed to have a gap of $\Delta_c = 2\Delta = 5$ meV, i.e., exactly twice the spin-wave gap at the "magic" reciprocal-space point. In Fig. 3 the result of this global fit is shown in a solid line, and the continuum contribution is represented by the dashed line.

The fact that the continuum starts above a well-defined gap energy $\Delta_c$, can be clearly seen in the wide-range constant-$q$ scans shown in Fig. 4. At this wave vector there are to spin-wave peaks due to a non-trivial 3D structure factor of the slightly zig-zag spin chains in $BaCu_2Si_2O_7$ (shaded areas). At high energies there is additional broad scattering not accounted for by the single-particle picture. The onset of the continuum is signaled by an intensity dip at around 5 meV (arrows). As in Fig. 3, the solid lines in Fig. 4 represent the global fit, and the dashed line is the continuum part of the

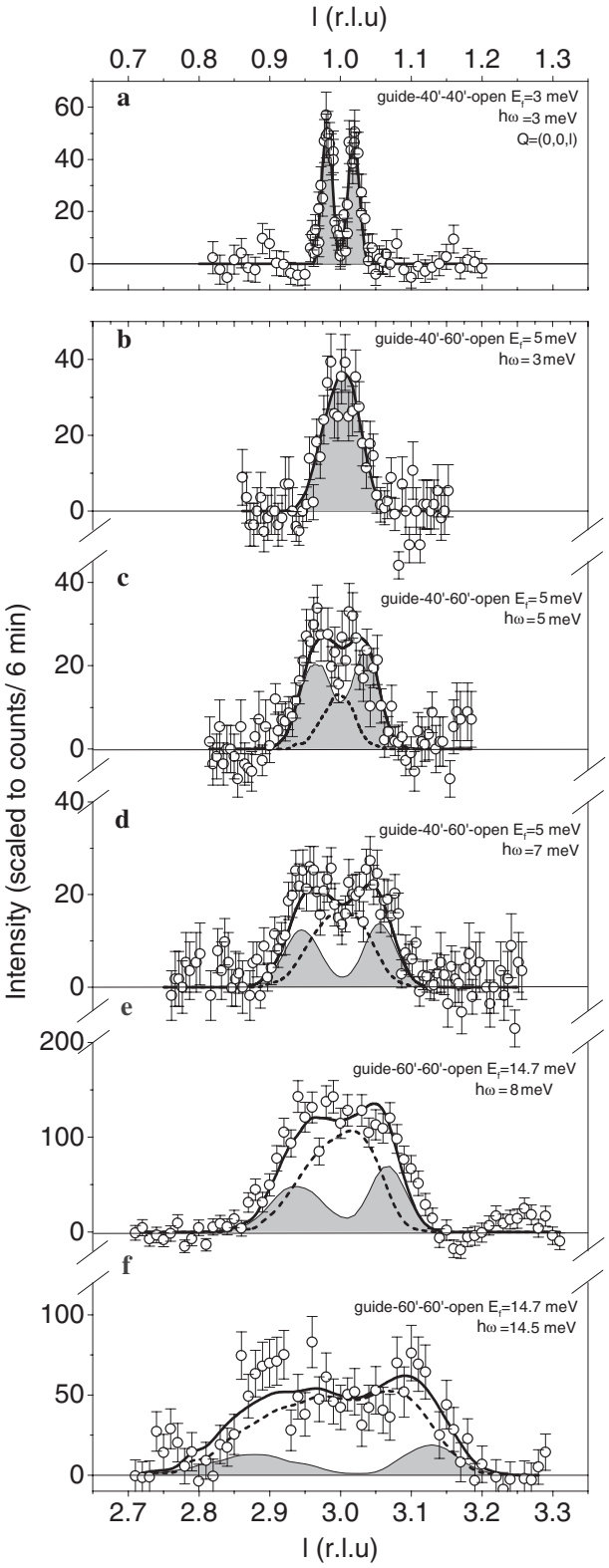


**Fig. 3.** A series of constant-$E$ scans along the spin chains in $BaCu_2Si_2O_7$. *Heavy solid* lines represent a global fit to the data as described in the text. *Shaded areas* are contributions of single-particle excitations. *Dashed lines* show the continuum portion. The data are from [32]

cross section. If $\Delta_c$ is treated as an adjustable parameter in the fit, the refined value is $\Delta_c = 4.8(2)$ meV, which is within the error bar of the theoretical value $\Delta_c = 2\Delta$ meV.

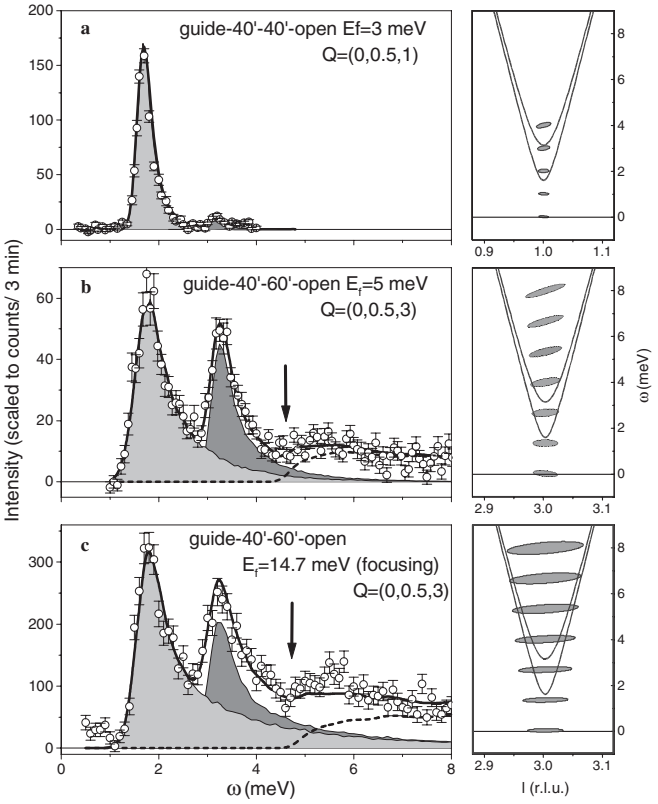


**Fig. 4.** *Left*: Typical constant-$Q$ scans collected in $BaCu_2Si_2O_7$ at the 1D AF zone center by using different spectrometer configurations. *Lines* and *shaded areas* are as in Fig. 3. *Arrows* indicate the slight dip in the observed intensity that corresponds to the continuum energy gap $\Delta_c$. *Right*: Evolution of the calculated FWHM resolution ellipsoids in the course of the corresponding scans, plotted in projection onto the $(l, \hbar\omega)$ plane. *Solid lines* represent the spin-wave dispersion relation. The data are from [32]

The continuum gap being *twice* the spin-wave gap is a non-trivial result. All the data discussed above were collected with scattering vectors almost parallel to the chain axis. The ordered moment in $BaCu_2Si_2O_7$ is parallel to the chains as well, so the intrinsic polarization dependence of the neutron scattering cross section ensures that all scans represent *transverse*-polarized spin fluctuations. In conventional SWT the lowest-energy transverse continuum excitations are *three*-magnon states, since the magnons themselves are transverse-polarized. In the SWT, the transverse continuum thus has a pseudogap of $3\Delta$. A rigorous SWT calculation for $BaCu_2Si_2O_7$ gives $\Delta_c = 7.5$ meV [31, 32]. How is it possible that we are seeing continuum scattering at $2\Delta$? The answer given by the quantum chain MF model is that since there are three possible polarizations for pairs of spinons $S_z = 0, \pm 1$ (see above), there is a *third* bound state (magnon) that is polarized parallel to the direction of ordered moment. In a recent, elegant study this *longitudinal mode* has been directly observed in $KCuF_3$ by using unpolarized [33] and polarized [34] neutrons. The longitudinal magnon is not visible in the $BaCu_2Si_2O_7$data shown above, due to polarization effects. However, it is the longitudinal mode that enables a *two*-magnon transverse-polarized continuum excitations with a gap $\Delta_c = 2\Delta$. Indeed, a transverse state can be constructed from one longitudinal and one transverse magnon. In other words, the fact that the continuum in $BaCu_2Si_2O_7$ starts at $2\Delta$ can

be taken as indirect evidence for the longitudinal mode. In the future it will be very important to perform neutron experiments in a different scattering geometry, perhaps using polarization analysis, to observe the longitudinal mode in $BaCu_2Si_2O_7$ directly, to corroborate the remarkable results on $KCuF_3$.

In summary, the seemingly simple model of weakly interacting spin chains demonstrates such fundamenatal phenomena of many-body quantum mechanics as mass generation, spinon confinement, and energy separation of "classical" and "quantum" spin dynamics. Studies of $KCuF_3$ and $BaCu_2Si_2O_7$ shed light on the nebulous regime where 1D quantum physics meets 3D "classical" magnetism and provide the experimental basis for some very sophisticated theoretical studies.

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