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TOPICAL REVIEW

Quantum and classical dynamics in mixed-spin one-dimensional antiferromagnets

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Abstract
Mixed-spin systems composed of quantum spin chains with gapped magnetic excitations interacting through 'auxiliary' magnetic ions show an effective separation between low-frequency classical and high-frequency quantum spin correlations. This phenomenon is realized in a family of rare-earth nickelates. Studies of these materials enable experimental measurements of some previously inaccessible fundamental properties of quantum spin chains to be made. The non-trivial intrinsic dynamics of the auxiliary spins gives rise to peculiar excitations of a mixed nature, and in certain cases may play a key role in long-range magnetic ordering.

1. Introduction

For the past two decades one-dimensional (1D) quantum antiferromagnets (AFs) have been among the hot topics in condensed matter physics. Of particular fundamental interest is the 1D Heisenberg AF that, despite the simplicity of its Hamiltonian, demonstrates some very non-trivial quantum-mechanical properties. Its classical ground state is the two-sublattice Néel state [1, 2], characterized by long-range magnetic order (LRO), a breaking of translational and rotational symmetries, and thus a large degeneracy. The Néel state is actually *not* an eigenstate of the quantum Heisenberg Hamiltonian [2]. In three dimensions corrections to the Néel state from zero-point quantum fluctuations can be taken into account perturbatively. However, in the 1D model these fluctuations totally destroyed LRO even at $T = 0$. The actual ground state is nothing like the two-sublattice state and is, in fact, not degenerate. Moreover, Haldane [3] showed that spin correlations are radically different in integer-spin and half-integer-spin systems. In particular, for integer-spin chains, equal-time spin correlations decay exponentially with distance. Such systems are truly disordered and are often referred to

as 'quantum spin liquids'. Their excitation spectrum is isotropic, and the low-energy excited states are triplets of massive particles. This is in stark contrast with the classical spectrum that consists of a pair of massless spin waves, polarized perpendicular to the direction of the ordered moment.

The wealth of experimental and theoretical results accumulated to date adds up to a fairly complete understanding of many of the properties of idealized 1D quantum AFs. One remaining problem of enormous interest is understanding the crossover from quantum-mechanical to classical behaviour that occurs in *quasi*-1D systems: interacting quantum spin chains as part of *real* three-dimensional crystal structures. Significant insight in this field has been obtained in studies of simple compounds with *directly coupled* quantum spin chains, such as $CsNiCl_3$ ($S = 1$) [4] and $KCuF_3$ ($S = 1/2$) [5]. In the present paper we discuss the more complex case of quantum spin chains that interact with each other only indirectly, via an 'auxiliary' network of essentially classical spins.

The only known experimental realizations of this model are rare-earth nickelates with the general formula R_2BaNiO_5. These 'mixed' quantum–classical systems contain almost isotropic $S = 1$ AF Ni chains weakly coupled to magnetic rare-earth ions. As discussed below, they exhibit an effective *separation* of classical and quantum spin dynamics in certain regimes. This provides a unique opportunity to investigate a number of interesting phenomena, including the effect of strong staggered magnetic fields on Haldane spin chains, and the peculiar interaction between collective quantum excitations in the chains and single-ion quantum transitions in the rare-earth elements.

All R_2BaNiO_5 species are derivatives of Y_2BaNiO_5, an isomorphous compound in which the magnetic R^{3+} are replaced by non-magnetic Y^{3+} ions. Y_2BaNiO_5 is a textbook example of a Haldane-gap AF [6]. Its magnetic properties are due to chains of $S = 1$ Ni^{2+} ions that run along the *a*-axis of the orthorhombic crystal structure (figure 1). The Ni spins are coupled by rather strong ($J \sim 300$ K) AF nearest-neighbour intra-chain interactions. Inter-chain coupling is negligible. Down to very low temperatures this system shows no sign of magnetic ordering and has a thermally activated magnetic susceptibility: a signature of a spin-singlet ground state and energy gap. Haldane excitations in Y_2BaNiO_5 (Haldane gap $\Delta \approx 10$ meV) have been observed and extensively studied in inelastic neutron scattering experiments [6]. In R_2BaNiO_5 compounds, the place of Y^{3+} is taken by magnetic rare-earth ions ranging from Pr^{3+} to Tm^{3+} [7], and recently Yb^{3+} [8]. Even though direct interactions between the R^{3+} moments are negligible, their coupling to the Ni chains in all cases leads to long-range Néel ordering with transition temperatures T_N ranging from 8.8 K (Yb_2BaNiO_5) to 65 K (Dy_2BaNiO_5). A static ordered moment appears on both the Ni and R sites simultaneously upon cooling through T_N (figures 2(a), 2(b)). At first glance it may seem that magnetic LRO is a clear sign of a destruction of the singlet ground state in individual Haldane chains. Indeed, for quite some time R_2BaNiO_5 compounds were considered to be classical magnets. A breakthrough came when, in inelastic neutron scattering experiments on Pr_2BaNiO_5 [9], Nd_2BaNiO_5 [10–12], and $(Nd_xY_{1-x})_2BaNiO_5$ [11, 13], it was discovered that 1D Ni-chain gap excitations, strikingly similar to the Haldane modes in Y_2BaNiO_5, not only exist above the ordering temperature, but also *persist in the magnetically ordered state*. Somehow the 1D quantum spin correlations in the Haldane chains survive the onset of LRO.

2. Two frequency regimes

The key to resolving this apparent contradiction lies in the very short timescale associated with dynamic spin correlations in individual chains, which in turn is a *direct consequence of the energy gap*. At frequencies substantially smaller than the threshold frequency $\Delta/\hbar$,

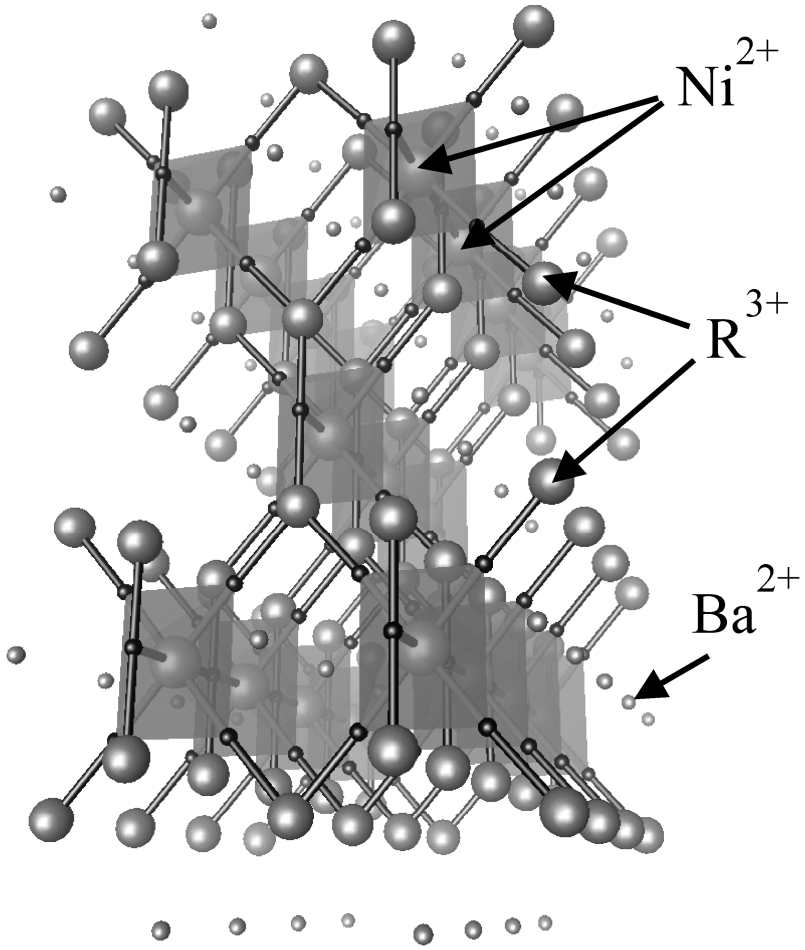

Figure 1. A schematic view of the R_2BaNiO_5 crystal structure. $S = 1$ Haldane spin chains are formed by Ni^{2+} ions at the centres of NiO_6 octahedra, arranged along the crystallographic *a*-axis. The rare-earth ions (spheres) provide magnetic links between chains.

the dynamic susceptibility $\chi(\omega)$ of an isolated $S = 1$ AF chain is purely real and is *only weakly frequency dependent*. In R_2BaNiO_5 the magnitude $J_\perp$ of Ni–R exchange interactions is typical of a rare-earth–transition-metal superexchange bond, and is of the order of 1 meV. This energy scale is much smaller than the Haldane gap Δ (10 meV). The local magnetization of the Ni chains is able to instantaneously follow any fluctuations of neighbouring rare-earth spins. At low frequencies the effect of the quantum spin chains is then reduced to simply providing an effective pathway for R–R interactions of the magnitude $J_{\mathrm{eff}} \sim J_\perp^2 \chi(0)$. We end up with a 3D network of essentially classical rare-earth moments that, not surprisingly, orders magnetically at low temperatures. The low-frequency dynamics is then also that of a classical magnet. Conversely, the same arguments suggest that the dynamic spin correlations in the high-frequency range are those of a 1D $S = 1$ quantum AF in a *static* effective field generated by the rare-earth subsystem. In other words, in the limit of weak coupling, for two-component systems that include *gapped* quantum spin chains there is a *separation* between static and low-frequency properties (3D LRO and conventional spin waves) and the high-frequency behaviour

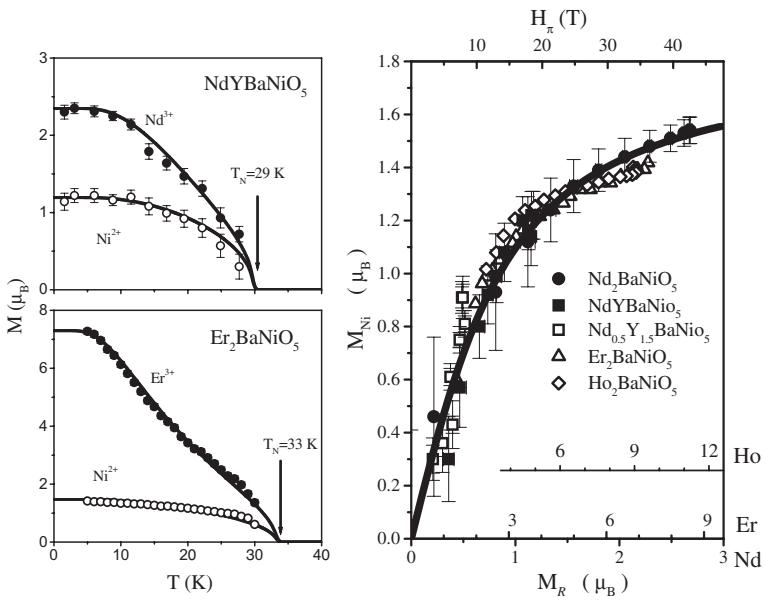

Figure 2. (a) Temperature dependences of the Ni- and R-sublattice magnetizations measured in $Nd_2YBaNiO_5$ (symbols) [11]. The solid lines are chain–MF theoretical fits. (b) The same for Er_2BaNiO_5 [23]. (c) The measured induced staggered moment on the Ni sites (symbols) plotted against the magnetic order parameter of the R sublattice in several R_2BaNiO_5 compounds (bottom axes). The collapsed data sets can be interpreted as a measurement of the universal staggered-magnetization curve for Haldane spin chains (top axis) [17]. The solid line is a theoretical prediction based on the ϕ^4-model [18].

(quantum 1D excitations). In the following sections, we shall review the experimental and theoretical results that support this physical picture.

2.1. Static properties

A quantitative model that embodies the hand-waving arguments given above is the chain–mean-field (chain–MF) theory [14, 15]. The standard MF approach for conventional magnets starts out with the bare (non-interacting) susceptibilities for all individual magnetic ions involved. In the chain–MF theory, one respects the fact that Ni–R coupling is substantially smaller than the strong interactions within the Ni chains themselves. The chains are thus treated as single entities. For the R_2BaNiO_5 series the low-temperature magnetic structure is antiferromagnetic, so the relevant properties of the spin chains are their *staggered* susceptibility χ_π and *staggered-magnetization function* $M_\pi(H_\pi)$. As for the rare-earth ions, for those with a Kramers doublet ground-state configuration (Nd^{3+} and Er^{3+} among them) it is appropriate to take the standard approach and write the non-interacting response in the form of a Brillouin function for an isolated ion. The coupling between these two systems is then treated at the MF level and all static properties, including long-range ordering and the T-dependence of the magnetic order parameters, are derived from a set of self-consistent equations. The problem is that to solve these equations one has to know $M_\pi(H_\pi)$ for an isolated chain. Prior to the experimental studies of R_2BaNiO_5 this function was not known analytically or even numerically. Simple powder-diffraction measurements on $(Nd_xY_{1-x})_2BaNiO_5$, Er_2BaNiO_5, and Ho_2BaNiO_5, however, allowed the direct *measurement* of this fundamental property [11,

17]. The magnetic ordering temperatures (around 50 K) are considerably smaller than the Haldane-gap energy ($\Delta/k_B \sim 120$ K) and, in this regime, $M_\pi(H_\pi)$ is almost T-independent [16]. The effective staggered field acting on the spin chains in R_2BaNiO_5 is proportional to the rare-earth magnetic order parameter. From these two facts it follows that $M_\pi(H_\pi)$ may be obtained simply by plotting the measured ordered moment on the Ni sites versus that on the R sites, as shown in figure 2(c). The scaling of the abscissa depends on the MF coupling constant and obviously depends on the type of the rare-earth ions involved. Universal scaling in proper magnetic field units is achieved by substituting the measured $M_\pi(H_\pi)$ curve into the self-consistent MF equations. The MF coupling constants are then refined by fitting the measured temperature dependencies of sublattice magnetizations [11, 17]. Excellent fits can be obtained in this manner (figures 2(a), 2(b)—solid lines). The resulting field scale is shown on the top axis of figure 2(c). The data collapse for systems with substantially different ordering temperatures and saturation moments is quite impressive, validating this approach. Note that staggered fields of up to 40 T are being produced in these experiments!

The direct measurements of the staggered-magnetization curve $M_\pi(H_\pi)$ for a Haldane spin chain stimulated new theoretical studies. A particularly successful approach [11, 18] is the O(3)-symmetric (1 + 1)-dimensional field theory (the so-called ϕ^4-model), first used by Affleck to describe Haldane spin chains [15]. In this theory it becomes apparent that the non-linearity of $M_\pi(H_\pi)$ is a manifestation of *repulsion* between gap excitations in the chains. Coefficients characterizing this repulsion can be estimated using RG theory [18] and used to obtain power series expansions for the staggered-magnetization curve. These predictions are in excellent agreement with the all the R_2BaNiO_5 data obtained to date and are shown as a solid line in figure 5(b)—see later.

It is important to realize that the mechanism of magnetic ordering in R_2BaNiO_5 materials is quite different from that in systems with directly coupled chains, such as $CsNiCl_3$. In the latter, LRO results from a complete softening of the Haldane excitations at the 3D AF zone centre as $T \to T_N$. This can happen only if inter-chain interactions exceed some critical value [15]. In the case of rare-earth nickelates, magnetic ordering is expected to occur for any *arbitrarily small* Ni–R coupling, as a result of the T^{-1}-divergence in the bare susceptibility of individual rare-earth moments.

2.2. Low-frequency spin dynamics

While long-range magnetic ordering is beautifully described by the chain–MF model, the spin *dynamics* in our two-component magnets can be analysed with the chain random-phase approximation (RPA). At this level, just as was the case for the static properties, the low-energy magnetic excitations in R_2BaNiO_5 can be described in terms of effectively coupled classical rare-earth spins. Such classical spin waves were recently studied in Nd_2BaNiO_5 [11]. The Nd case is particularly simple because of a strong easy-axis magnetic anisotropy associated with Nd^{3+}. The spin waves are then dispersionless and resemble excitations in a conventional Ising magnet (figure 3, inset). The excitation energy is given by $2M_{Nd}H_\pi$, where M_{Nd} is the saturation moment of Nd^{3+} and H_π is the effective mean field. In fact, the measured temperature dependence of this energy (figure 3 main panel, symbols) can be accurately reproduced, without using any adjustable parameters, by utilizing $H_\pi(T)$ determined from the MF analysis of the sublattice magnetization data (figure 3 main panel, solid line).

2.3. High frequencies

We now turn to the gap excitations in the Ni^{2+} chains. In the paramagnetic phase (above T_N) there is no ordered moment on the rare-earth sublattice, so the 1D gap modes are exactly

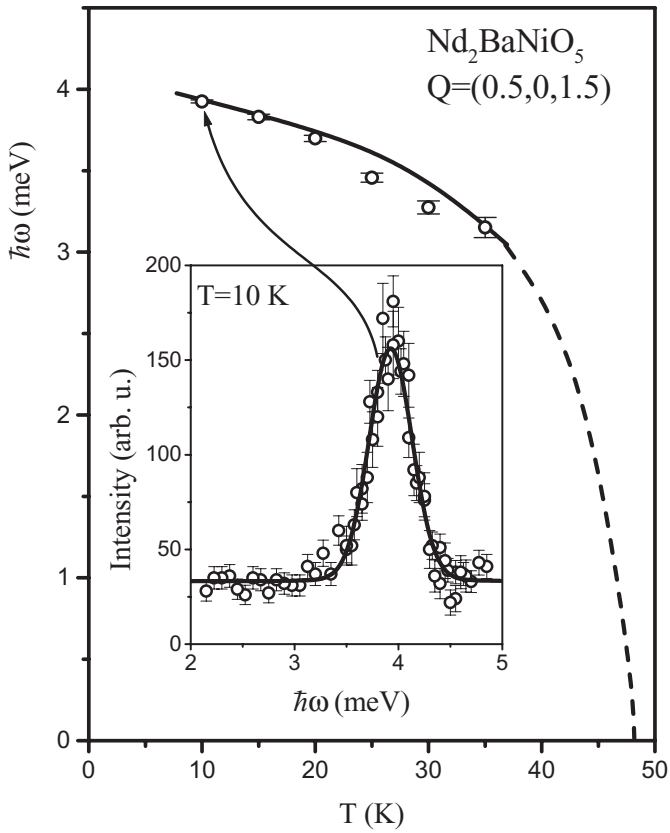

Figure 3. Order-parameter excitation (spin wave) in Nd_2BaNiO_5 observed for constant Q at $T = 10$ K (inset). This dispersionless Ising-like excitation is observed only in the ordered state and its energy (main panel, symbols) is proportional to the effective staggered field in the system (solid and dashed lines). The data are from reference [11].

as those in uncoupled chains. Thus in Nd_2BaNiO_5 and Pr_2BaNiO_5, for $T > T_N$, the 1D gap excitations are experimentally identical to those found in Y_2BaNiO_5 [9, 11–13]. In particular, the T-dependence of the gap energy is very similar (figure 5(a)—see later). Within the framework of our MF–RPA model, in the magnetically *ordered* state, the gap modes are those of isolated quantum spin chains immersed in a *static* exchange field generated by the ordered rare earths. This simple observation turns R_2BaNiO_5 compounds into unique model systems for fundamental studies of quantum spin chains in strong, adjustable staggered fields.

Since a staggered field is the conjugate of the classical order parameter, it will induce a crossover from quantum to classical spin dynamics in individual chains. The spectrum of a classical Heisenberg AF is twofold degenerate, while that in the quantum $S = 1$ model contains three magnon branches. The first effect that we can expect a staggered field to have on a Haldane spin chain is to suppress the 'extra' longitudinal mode. In the presence of a staggered field, the classical AF develops a gap that increases linearly with H_π for large H_π. In the quantum system an increase of the gap energy can be expected as well. As the Haldane spin chain is gapped even at $H_\pi = 0$, the change of the gap energy should start out quadratic with H_π and cross over to a linear dependence for large staggered fields, merging with classical behaviour.

Polarized-neutron studies of Nd_2BaNiO_5 demonstrate how this crossover actually occurs [12]. Figure 4 shows background-subtracted scans collected in this compound below and above the ordering temperature. Solid symbols correspond to transverse excitations, i.e., the contribution to the magnetic dynamic structure factor from spin components perpendicular to the direction of the ordered moment in the low-temperature phase. Open symbols represent longitudinal spin fluctuations. At $T > T_N$, in the absence of a staggered field, the observed scattering is indeed isotropic (figure 4(a)). However, at $T < T_N$, when the staggered field is turned on, the intensity of the 'forbidden' longitudinal mode decreases (figure 4(b)). At still lower temperatures (higher staggered fields) the longitudinal component practically disappears. The non-trivial result is that even in the ordered phase, the mode polarized along the direction of staggered magnetization, and thus *totally absent* from the conventional spin-wave theory, persists as an underdamped excitation.

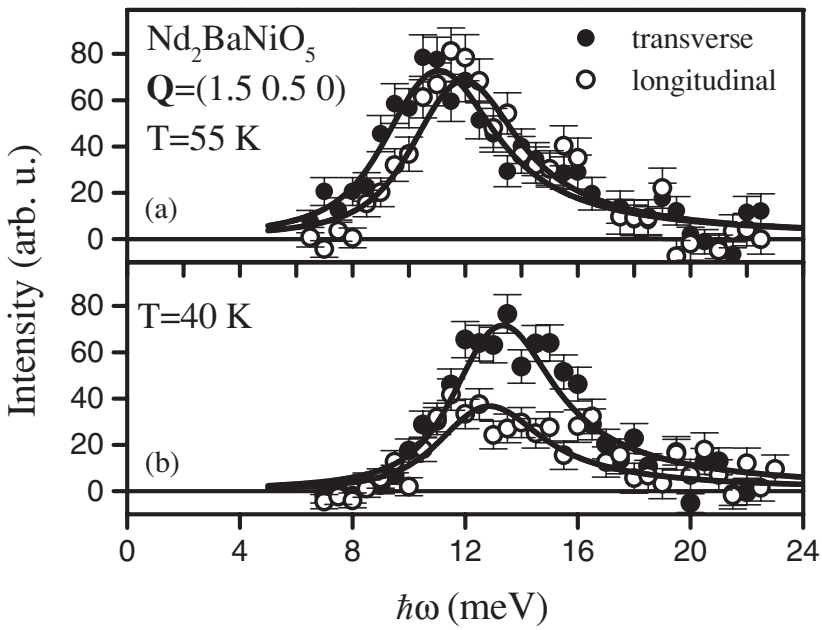

Figure 4. Constant-Q scans collected at the 1D AF zone centre in Nd_2BaNiO_5 showing the Ni-chain gap excitations in the paramagnetic (a) and ordered (b) phases. At $T > T_N = 48$ K the spectrum is isotropic (a). Below T_N (b) the intensity of the mode polarized along the direction of the ordered moment (open symbols) decreases, while that of the transverse mode (solid symbols) remains unchanged. The gap increases with decreasing temperature in both channels of spin polarization. The data are from reference [12].

Once the relation between temperature and staggered field in R_2BaNiO_5 has been determined from the analysis of sublattice magnetizations, the staggered-field dependence of the gap energy can be directly measured. As seen in figure 5(a), Δ increases with decreasing T below T_N, roughly linearly with $T - T_N$. The behaviour is independent of the type of rare earth involved or the actual Néel temperature. The best way to demonstrate this universality is to eliminate the rare-earth-related energy scale completely by plotting the increase of the Haldane gap, normalized to that in Y_2BaNiO_5, as a function of the induced static staggered moment on the Ni sites (figure 5(b)). When plotted in this way, the data collapse onto a single curve, revealing the expected quadratic behaviour. Like the experimental staggered-magnetization curve, the measured staggered-field dependence of the gap energy is in excellent quantitative

agreement with recent theory. The solid line in figure 2(c) is the ϕ^4-result, drawn over the data with no adjustable parameters [11, 18]. Density matrix renormalization group calculations [19] are shown by a dashed line. The increase of the gap energy in a two-component magnet was recently rigorously proven for the valence-bond-solid model (VBS) [20], known to possess many similarities with the 1D $S = 1$ Heisenberg model relevant to the present case. A more detailed σ-model analysis of the Heisenberg version of the model was also carried out [21].

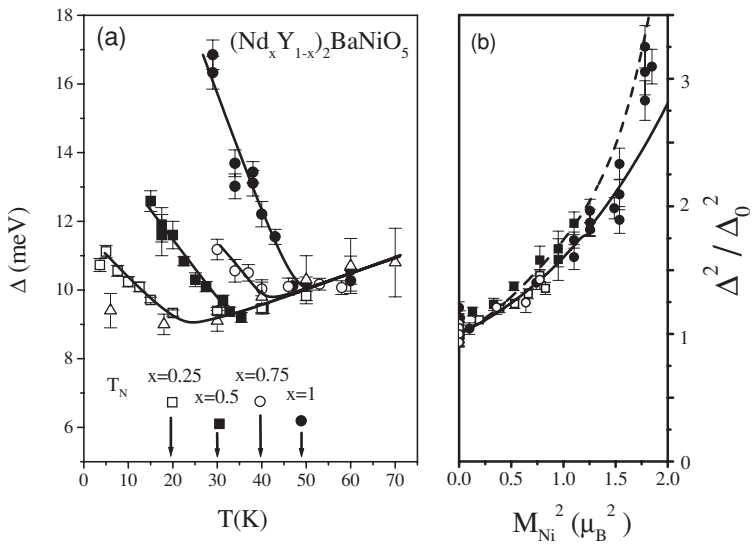

Figure 5. (a) The measured temperature dependence of the Haldane-gap energy for a series of Y-substituted Nd_2BaNiO_5 compounds (symbols) [11, 13]. Above the corresponding temperatures of magnetic ordering all systems show the same behaviour as the intrinsically disordered Y_2BaNiO_5 [6]. In the ordered phase the gap increases in all cases linearly with T. Lines are guides for the eye. (b) The data collapse obtained by plotting the measured increase of the gap in $(Nd_xY_{1-x})_2BaNiO_5$ relative to that in Y_2BaNiO_5 as a function of the induced staggered moment on the Ni chains. The solid line is a parameter-free prediction of the ϕ^4-model [18], and the dashed line represents recent numerical results [19] for Haldane chains in a staggered field.

3. Crystal-field excitations

Above, we restricted ourselves to considering Haldane chains coupled to essentially classical spins. Qualitatively new phenomena occur when the 'auxiliary' magnetic ions themselves have a non-trivial intrinsic dynamics. Due to strong spin–orbit interactions and low site symmetry, this in fact is always the case for R^{3+} in R_2BaNiO_5. Not just the ground-state multiplets of the rare-earth ions, but also their higher-energy excited crystal-field (CF) configurations should thus be considered. Here one can identify two distinct regimes. The first is realized when the R-centred single-ion CF transitions occur close to, or above, the quantum gap in the Ni chains. CF excitations then have little influence on the low-frequency and static properties of the system. Instead, as will be illustrated below, their proximity to the Haldane modes gives rise to peculiar mixed Ni–R excitations. In the second regime the energies of the relevant excited CF states are considerably smaller than the Haldane gap, and are comparable to the magnitude of the Ni–R interactions. CF transitions in this case have a strong influence

on the low-frequency dynamics and may become the driving force of long-range magnetic ordering.

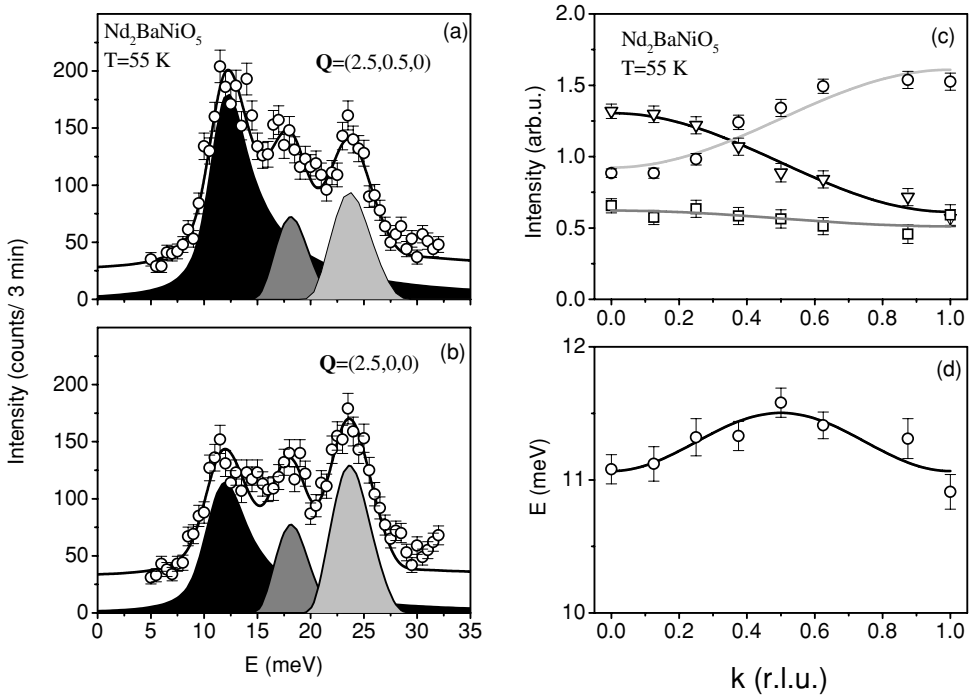

Figure 6. Mixing between Haldane-gap and crystal-field excitations in Nd_2BaNiO_5. ((a), (b)) Typical constant-Q scans measured at the 1D AF zone centre for two different momentum transfers perpendicular to the chain axis (symbols). The heavy solid line is a fit to a model cross-section taking into account resolution effects. The black, dark grey and light grey shaded peaks represent the Haldane and two CF excitations, respectively. This analysis reveals a substantial intensity modulation in all modes (c) and a weak dispersion in the Haldane mode (d).

3.1. High-frequency CF excitations

The first regime is known to be realized in Nd_2BaNiO_5 [10]. The interactions between Ni-chain and Nd-single-ion excitations in this material were recently investigated by means of single-crystal inelastic neutron scattering [25]. Representative constant-Q scans collected at the 1D AF zone centre for different momentum transfers perpendicular to the chain axis, at $T = 55$ K (*above* $T_N = 48$ K), are shown in figures 6(a), 6(b). Three separate peaks, namely the Haldane-gap excitation at 11 meV energy transfer and two CF excitations, a weaker mode at 18 meV and a strong one at 24 meV, are observed. A striking feature is the strong variation of the intensity of these modes as a function of transverse momentum transfer. A quantitative analysis of this variation was performed by fitting individual scans to a model cross-section convoluted with the four-dimensional spectrometer resolution function, as shown in figures 6(a), 6(b) as solid black lines. This procedure extracts the b-axis modulation of the energy-integrated intensity for each mode (figure 6(c)). In addition, it revealed a very weak transverse dispersion in the Haldane branch (figure 6(d)). No dispersion in the CF excitations could be detected with the relatively poor energy resolution at high energy transfers ($\approx$3.5 meV FWHM at 20 meV). A mesh composed of data points collected in constant-E scans

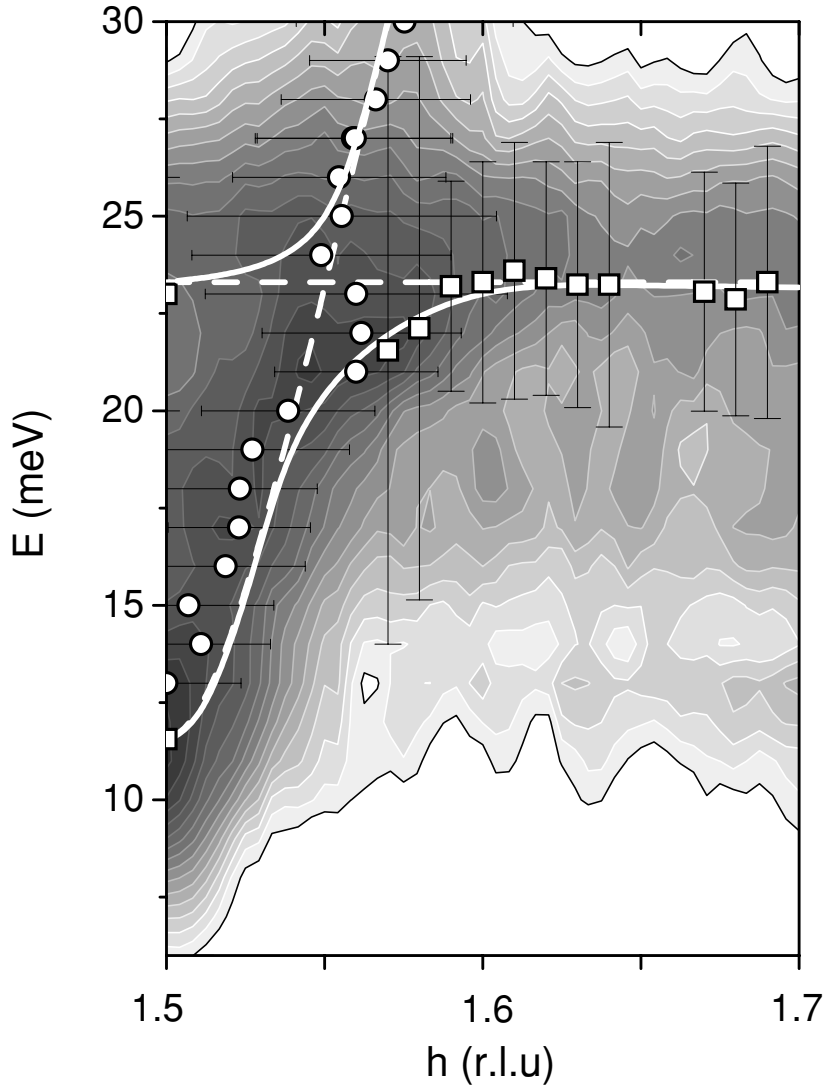

Figure 7. A greyscale plot of inelastic neutron scattering intensity measured in Nd_2BaNiO_5 at the 1D AF zone centre, which shows the highly dispersive Haldane-gap mode (parabola) and the flat 24 meV crystal-field mode (horizontal dashed line). The symbols and attached bars show the positions and widths of peaks seen in constant-E (circles) and constant-Q (squares) scans, respectively. A mixing between the two modes leads to a mutual repulsion at the point of their intersection (solid lines).

along the chain axis is shown in figure 7 and reveals an even more significant result, namely an 'anticrossing' of the Haldane and the 24 meV CF excitations.

The observed intensity modulation and weak transverse dispersion, as well as mode repulsion, are a clear sign of a mixing between Ni-chain Haldane-gap modes and single-ion R-centred CF excitations. This behaviour is well accounted for by a slightly more sophisticated version of the chain–RPA model, that includes higher-energy levels of the rare-earth ions, as discussed in reference [25]. Note that the 1D AF zone centre is sufficiently far away from the points of anticrossing, and the effect of the weak Ni–R coupling on the *energy*

of the 1D gap modes is rather small (of the order of $(J_\perp/J)^2$), so the interpretation of the T-dependence of the gap in terms of Haldane chains in a static staggered field is still justified. The *intensity* modulation however is substantial, being linear with $(J_\perp/J)$. 1D gap excitations in Nd_2BaNiO_5 can therefore be described as Haldane modes propagating on the Ni chains, 'dressed' by CF fluctuations on the Nd sites.

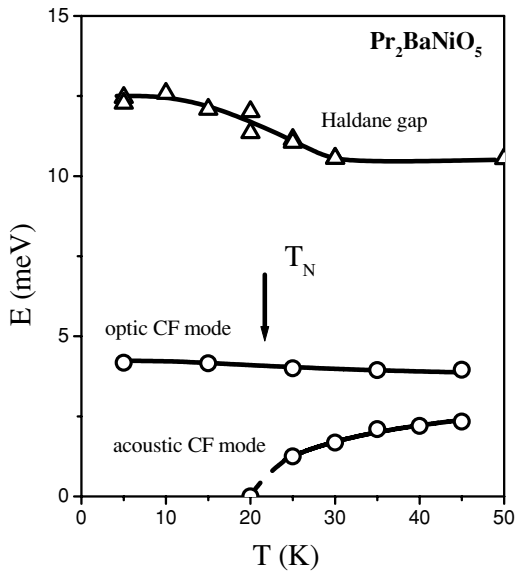

Figure 8. The temperature dependence of the 3D AF zone-centre excitation energies measured in Pr_2BaNiO_5 [9]. A repulsion between single-ion Pr CF excitations (circles) and Haldane-gap modes in the Ni chains (triangles) leads to a soft-mode transition to a Néel-ordered state below $T_N = 24$ K, despite the singlet (non-magnetic) nature of the Pr^{3+} ions. The Haldane-gap modes behave very similarly to those in Nd_2BaNiO_5.

3.2. Low-frequency CF excitations

Quite different is the second regime, where the energies of certain excited CF states are considerably smaller than the Haldane gap. As in the case of classical 'auxiliary' spins, rare-earth and Ni-chain spin dynamics are effectively separated. At low frequencies the Haldane chains simply provide effective pathways for direct R–R interactions. This effective coupling becomes particularly important for non-Kramers rare-earth ions that in the structure of R_2BaNiO_5 have a non-magnetic ground state. A good example is Pr_2BaNiO_5, where Pr^{3+} has a spin-singlet ground state, but in addition has a CF-excited state at only 4 meV energy. Above $T = 100$ K the corresponding CF mode is dispersionless. With decreasing temperature it acquires a substantial dispersion and intensity variation both along, and perpendicular to, the chain axis [9]. With two Pr sites per Ni site, The CF mode becomes split into optical and acoustic branches. As shown in figure 8, the acoustic branch develops a sharp dip at the 3D AF zone centre, where its energy reaches zero at $T_N = 24$ K. A magnetic *soft-mode* transition occurs at this point, and is driven by Ni-chain-mediated Pr–Pr interactions. This soft-mode behaviour is very similar to that in metallic Pr [24]. Below T_N, Pr_2BaNiO_5 is ordered in a structure similar to that of Nd_2BaNiO_5. In an unusual twist, magnetic LRO in this case is a result of interaction between two

non-magnetic systems: singlet-ground-state rare-earth ions and quantum-disordered spin chains.

4. Summary

In summary, complex systems composed of gapped quantum spin chains interacting through isolated free spins demonstrate a rich spectrum of magnetic properties with both classical and quantum-mechanical features. R_2BaNiO_5 rare-earth nickelates are the unique model compounds for which these phenomena can be studied experimentally.

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