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High-field spin dynamics of antiferromagnetic quantum spin chains

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Abstract

The characteristic internal order of macroscopic quantum ground states in one-dimensional spin systems is usually not directly accessible, but reflected in the spin dynamics and the field dependence of the magnetic excitations. In high magnetic fields quantum phase transitions are expected. We present recent work on the high-field spin dynamics of the $S=1$ antiferromagnetic Heisenberg chains NENP (Haldane ground state) and $CsNiCl_3$ (quasi-1D HAF close to the quantum critical point), the uniform $S=\frac{1}{2}$ chain CTS, and the spin-Peierls system $CuGeO_3$. © 2000 Elsevier Science B.V. All rights reserved.

Keywords: One-dimensional systems; Haldane gap; Peierls transition; Antiferromagnets

Quasi-one-dimensional spin systems with spin values $\frac{1}{2}$ and 1 display a rich variety of macroscopic ground states, among which the collective singlet ground states have attracted particular attention. The Haldane ground state of the $S=1$ Heisenberg antiferromagnetic chain is characterized by a non-local antiferromagnetic order. It has been found to be stable for a certain range of anisotropies, interchain interactions, and uniform magnetic field, because the excitations have a finite energy of $E \geqslant \Delta = 0.411(2J)$. At the critical field value $g\mu_B H_{c1} = \Delta$ the system enters a gapless phase, where the transverse antiferromagnetic correlation length diverges [1]. With a finite interchain interaction, and/or an orthorhombic anisotropy a long-range ordered Néel state is expected above H_{c1}. The upper critical field $g\mu_B H_{c2} = 8JS$ overcomes the intrachain interaction. According to calculations on finite chains [2], for $M \geqslant \frac{1}{2}M_{sat}$, incommensurate (IC) soft modes at $q \propto M - M_{sat}/2$ track through the Brillouin zone. Other authors [3] predict the IC soft modes already above $g\mu_B H_{c1}$, with $q \propto M$. We investigated the spin dynamics of the prototype Haldane system NENP at (1.1 1 0) with $H \| c$. The dispersion of the lowest excitation continues to flatten above the critical field [4,5], while the middle one regains the zero-field spin-wave velocity. Apart from this flattening of the lowest excitation there is no evidence of incommensurate modes (Fig. 1).

One might argue, that the forming of IC soft modes is inhibited by the staggered magnetic field, generated in the uniform field by the staggered components of the g-tensor. On the other hand, the observed staggered magnetization for $H \| c$, $H > H_{c1}$ definitely has a component parallel to a, as for a material with uniform g-tensor, whereas the staggered parts of the Ni g-tensor only produce staggered magnetization parallel to b for $H \| c$. That means, the antiferromagnetic-ordered moment

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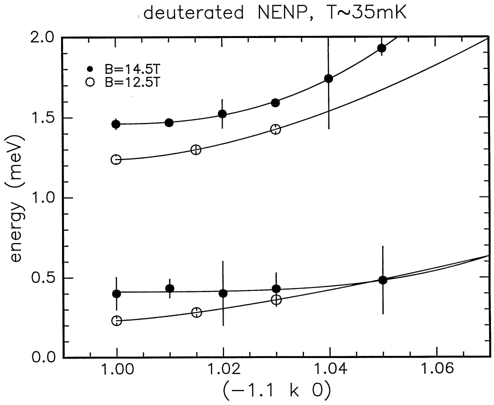

Fig. 1. Dispersion of the lowest two excitations of deuterated NENP along (1.1 k 0) in the high-field phase with a magnetic field $B \| c$ at $T \approx 35$ mK.

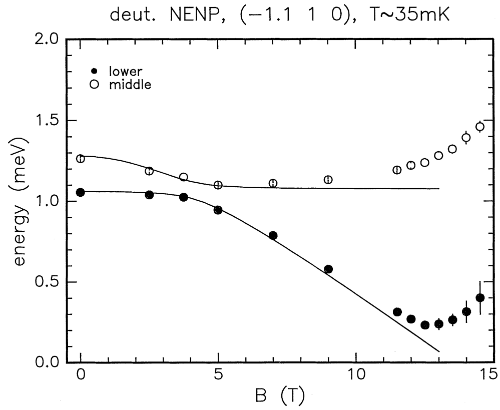

Fig. 2. Field dependence of the lowest two excitations of deuterated NENP at (1.1 1 0).

is not dominated by the staggered parts of g, therefore the spin dispersion can be expected to have similarities to that of a system with uniform g. The observed field dependence (Fig. 2) and polarization of the lowest two excitations shows that the sign of the orthorhombic anisotropy is reversed in the deuterated with respect to the protonated compound [6,5]. Above H_{c1}, the excitations follow roughly $g\mu_B H$. The polarization of the excitations changes again, leading to increasing c-contributions as expected for a transverse excitation. Nevertheless the results call for calculations of the spin-dynamics of the high-field phase which include the anisotropy, staggered field components and interchain interactions.

$CsNiCl_3$ is only close to the Haldane ground state as it orders antiferromagnetically at 4.8 K. Nevertheless it behaves like a Haldane system in the paramagnetic phase [7], and the spin-wave theory does not describe excitations of the long-range-ordered phase [8,9]. Additional sharp branches with a strong field dependence have been detected and are explained in a Lagrangian derived from the non-linear σ model [9]. These modes are now detected at the 1D AF zone center as well [5], where they could not be observed previously [8]. The results confirm, that Haldane ground state correlations survive even deep in the ordered phase of quasi-1D HAF systems.

The spin $\frac{1}{2}$ Heisenberg antiferromagnetic chain has gapless excitations with most of the spectral weight at the lower boundary of a continuum. In contrast to the classical HAF chain, incommensurable soft modes with $q \propto H$ are expected in a magnetic field and the lower boundary of $S_{\|H}(q,\omega)$ develops a dispersion which *decreases* from the gap $g\mu_B H$ at the 1D AF zone center [10]. We have observed these features in the paramagnetic phase of CTS [5].

The ground state of the $S = \frac{1}{2}$ HAF chain is instable against dimerization if an appropriate lattice distortion couples to the spin system. Above a critical magnetic field, the dimerized state of a spin-Peierls chain is broken and the structure becomes incommensurate. Incommensurate soft magnetic excitations are expected in the high-field phase [3,11]. We investigated the spin dynamics of $CuGeO_3$ in the high-field phase at the AF zone center $(0\,1\,\frac{1}{2})$ [12] and find no indication of excitation minima at incommensurable q. The triplet modes quickly disappear at the critical field. Two sharp low-energy modes at 0.6 and 1 meV, respectively, are found immediately above H_c, the 2 meV mode of the Peierls phase disappears in a continuum-like broad feature.

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