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Observation of two-magnon bound states in the spin-1 anisotropic Heisenberg antiferromagnetic chain system $NiCl_2{-}4SC(NH_2)_2$

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Abstract

Results of systematic tunable-frequency ESR studies of the spin dynamics in $NiCl_2{-}4SC(NH_2)_2$ (known as DTN), a gapped $S=1$ chain system with easy-plane anisotropy dominating over the exchange coupling (large-D chain), are presented. We have obtained direct evidence for two-magnon bound states, predicted for $S=1$ large-D spin chains in the fully spin-polarized (FSP) phase. The frequency–field dependence of the corresponding excitations was calculated using the set of parameters obtained earlier [S.A. Zvyagin, et al., Phys. Rev. Lett. 98 (2007) 047205]. Very good agreement between the calculations and the experiment was obtained. It is argued that the observation of transitions from the ground to two-magnon bound states might indicate a more complex picture of magnetic interactions in DTN, involving a finite in-plane anisotropy.

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Antiferromagnetic (AFM) quantum spin-1 chains have been the subject of intensive theoretical and experimental studies, fostered especially by the Haldane conjecture [1]. The presence of a strong easy-plane anisotropy D can significantly modify the excitation spectrum [2] so that the gap size is not determined by the strength of the AFM quantum fluctuations *exclusively*, but depends on the dimensionless parameter $\rho = D/J$ (where J is the exchange coupling). For $\rho > 0.93$ [3], the origin of the gap is dominated by the anisotropy D and the system is in the so-called large-D regime. The gap can be closed by a magnetic field, H_{c1}, applied perpendicular to the anisotropy plane. When the magnetic field exceeds the upper critical field, H_{c2}, the system is in the fully spin-polarized (FSP) state and the low-energy magnetic excitation spectrum is formed by magnons. In addition to ordinary single-magnon states, the theory [4] predicts the existence of two-magnon bound states. In this paper, we report a detailed frequency–field diagram of magnetic excitations in DTN, a system of weakly interacting $S=1$ chains with single-ion anisotropy D larger than the intra-chain exchange coupling J_c, obtained by means of the high-field electron spin resonance (ESR) technique [5].

The compound $NiCl_2{-}4SC(NH_2)_2$ (dichloro-*tetrakis* thiourea-nickel(II), known as DTN) has a tetragonal crystal structure with space group I4 [6]. The g-factor, anisotropy, intra- and inter-chain exchange parameters, $g_c = 2.26$, $D = 8.9\,K$, $J_c = 2.2\,K$, and $J_{a,b} = 0.18\,K$, respectively, were obtained from high-field ESR data [7], zero-field inelastic-neutron scattering, and magnetocaloric measurements [8]. At sufficiently low temperatures ($T < 1.2\,K$) and at $H \| c$ (where c is the tetragonal axis), DTN undergoes a transition into a field-induced AFM

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ordered phase with $H_{c1} = 2.16\,\mathrm{T}$ and $H_{c2} = 12.6\,\mathrm{T}$ (defined at $T \to 0$) and critical exponents consistent with the Bose–Einstein-condensation universality class [8,9].

At $H < H_{c1}$, DTN is in the quantum-paramagnetic phase, having an $S_z = 0$ ground state and a gapped excitation spectrum determined by the $\Delta S_z = \pm 1$ transitions (the modes A and B in Fig. 1). Excitations from the ground to single-magnon states (the mode C in Fig. 1) were observed at $T = 1.6\,\mathrm{K}$ in the FSP phase. These excitations correspond to a single spin flip from the $S_z = -1$ to 0 state and are uniformly delocalized over the entire lattice with a well-defined momentum $\mathbf{k}$. The ESR transitions taking place at $\mathbf{k} = 0$ have the frequency $\omega_C = g\mu_B H - D$. From this expression, the single-ion anisotropy in DTN can be calculated accurately ($D = 8.9\,\mathrm{K}$) [7].

The two-magnon bound states (sometimes referred to as single-ion bound states) were predicted in 1970 by Silberglitt and Torrance [10] for Heisenberg ferromagnets with single-ion anisotropy. Later on, this subject attracted a great deal of attention due to its potential relevance to the intrinsic localized spin modes in anisotropic ferromagnets [11] and antiferromagnets [12]. The two-magnon bound states for $S = 1$ AFM chains in the FSP phase have been predicted by Papanicolaou et al. [4]. The physical picture of the two-magnon bound-state excitations corresponds to a double-spin-flip transition from $S_z = -1$ to $+1$. Since the diagonal energy difference between these two states, $2D$, is much bigger than exchange interactions, the distance between the two $S_z = 0$ sites remains finite, giving rise to a two-magnon bound state. A first signature of two-magnon bound states was obtained by means of high-field ESR in the spin-1 chain compound $Ni(C_2H_8N_2)_2Ni(CN)_4$ (known as NENC) [14], which was interpreted as transitions from the single-magnon to two-magnon bound states.

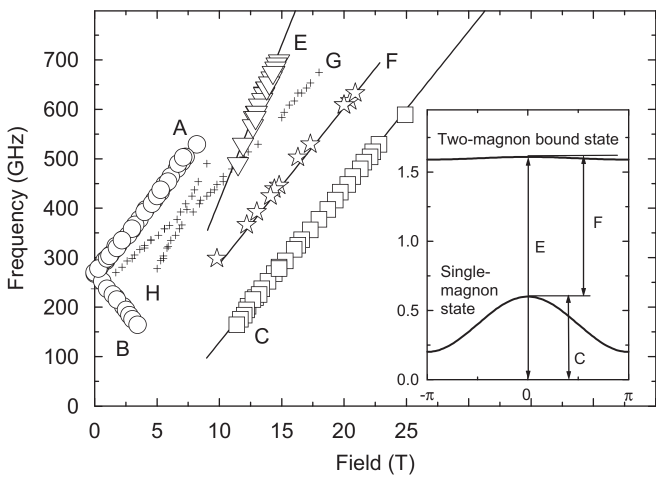

Fig. 1. Frequency–field dependence of magnetic excitations in DTN, taken at $H \| c$. Symbols denote the experimental results, and lines correspond to results of calculations (see text for details). The inset shows schematically the excitation dispersion in an $S = 1$ Heisenberg chain with strong easy-plane anisotropy in the FSP phase (note that the ESR transitions denoted by C, E, and F occur at $\mathbf{k} = 0$). The two-particle continuum is not shown for simplicity.

In this paper we report the direct observation of transitions from the ground state to the two-magnon bound state (the mode E, Fig. 1) in a spin-1 AFM chain system with strong easy-plane anisotropy in the high-field FSP phase. The corresponding excitations observed at $T = 1.6\,\mathrm{K}$ are denoted by triangles in Fig. 1 together with results of analytical calculations, using the set of parameters from Ref. [7] and the expression

$$\omega_E = -4(J_c + 2J_a) + 2J_c^2/D + 4J_a^2/D + 2g\mu_B H. \quad (1)$$

Very good agreement with experimental data was achieved. Since these excitations involve $\Delta S_z = 2$ transitions, which are strictly forbidden in case of an ideal axial symmetry [13], their observation by means of ESR might indicate that the axial symmetry in FSP phase is broken.

Observation of the modes G and H (Fig. 1) might indicate existence of a second $S = 1$ paramagnetic center in our samples. Although the exact origin of these modes is not clear at the moment, one can speculate that they are probably associated with excitations of Ni^{2+} spins in a superficial layer of DTN-crystals attacked by the GE-varnish solvent (used to fix the sample inside the sample-holder).

Finally, we observe a resonance absorption denoted by stars in Fig. 1. This ESR mode occurs at higher temperatures ($T = 4.3\,\mathrm{K}$) and results from transitions within excited states, i.e. between the single-magnon and two-magnon bound states at $\mathbf{k} = 0$. The frequency–field dependence of this transition (the mode F) can be calculated using the expression $\omega_F = \omega_E - \omega_C$ (where ω_E and ω_C are the excitation frequencies for the modes E and C, respectively).

In summary, an ESR study of the magnetic excitations in DTN, an $S = 1$ Heisenberg AFM chain material with strong single-ion anisotropy, has been presented. Two-magnon bound states, predicted for $S = 1$ large-D spin chains in the FSP phase, have been observed directly. The observation of $\Delta S_z = 2$ transitions from the ground to two-magnon bound states might indicate a more complex picture of the magnetic interactions in DTN, involving a finite single-ion in-plane anisotropy or the Dzyaloshinskii–Moriya interaction.

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