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Elementary excitations in $S = \frac{1}{2}$ Heisenberg spin chains with alternating g-tensor and the Dzyaloshinskii-Moriya interaction

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Abstract. The magnetic-excitation spectrum of copper pyrimidine dinitrate, a material containing $S = \frac{1}{2}$ antiferromagnetic chains with alternating g-tensor and the Dzyaloshinskii-Moriya interaction, and exhibiting a field-induced spin gap, is probed using tunable-frequency electron spin resonance spectroscopy in magnetic fields up to 25 T. The data are interpreted in frame of the sine-Gordon quantum-field theoretical concept proposed recently by Oshikawa and Affleck. The field-induced gap is measured *directly*; signatures of soliton and three breather branches are identified.

An isotropic $S = \frac{1}{2}$ Heisenberg antiferromagnetic (AF) chain with uniform nearest-neighbor exchange coupling represents one of the most remarkable models of quantum magnetism. Its ground state is a spin singlet, and the dynamics are determined by a gapless two-particle continuum of spin-$\frac{1}{2}$ excitations, commonly referred to as spinons. A uniform external magnetic field causes a substantial rearrangement of the excitation spectrum, making the soft modes incommensurate [1], although the spinon continuum remains gapless. Since the $S = \frac{1}{2}$ AF chain is critical, even small perturbations can considerably change fundamental properties of the system. One of the most prominent examples is the $S = \frac{1}{2}$ AF chain perturbed by an alternating g-tensor and/or the Dzyaloshinskii-Moriya (DM) interaction. In the presence of such structural peculiarities, the application of a uniform external field H induces an effective transverse staggered field $h \propto H$, which leads to the opening of an energy gap $\Delta \propto H^{2/3}$. It has been shown [2,3] that the gapped phase can be effectively described by the quantum-field sine-Gordon theory. The excitation spectrum consists of solitons, antisolitons, and multiple soliton-antisoliton bound states called breathers. The availability of exact solutions for the sine-Gordon model, allows very precise theoretical descriptions of many observable properties and physical parameters of sine-Gordon magnets (including the field dependence of excitation energies [2-4] and response functions [5,6]), which makes such systems a particularly interesting target for experimentally probing elementary excitations.

Here, we report on a detailed study of the low-temperature elementary excitation spectrum in copper pyrimidine dinitrate (hereafter Cu-PM), which has been recently identified as a $S = \frac{1}{2}$ AF chain with a field-induced spin gap [7], and is probably the best realization of the quantum sine-Gordon spin-chain model known to date. Cu-PM, $[\text{PM-Cu}(NO_3)_2(H_2O)_2]_n$ (PM = pyrimidine) crystallizes in a monoclinic structure belonging to the space group $C2/c$ with four formula units per unit cell. The lattice constants obtained from the single-crystal X-ray diffraction are $a = 12.404$ Å, $b = 11.511$ Å, $c = 7.518$ Å, and $\beta = 115.0°$. The Cu coordination sphere is a distorted octahedron, built from an almost square N-O-N-O equatorial plane and two oxygens in the axial positions. The Cu ions form chains running parallel to the short ac diagonal. The local principal axis of each octahedron is tilted from the ac plane by $\pm 29.4°$. Since this axis almost coincides with the principal axis of the g-tensor, the g-tensors for neighboring Cu ions are staggered. The exchange constant determined from susceptibility measurements [7] is $J = 36 \pm 0.5$ K.

The excitation spectrum was studied using a high-field submillimeter wave ESR spectrometer [8]. Backward Wave Oscillators were employed as tunable sources of radiation, quasi-continuously covering the frequency range of 150 to 700 GHz. The magnetic field was applied along the c'' direction, which is characterized by the maximal value of the staggered magnetization for Cu-PM [7]. The experiments were performed both in the Faraday and the Voigt configurations.

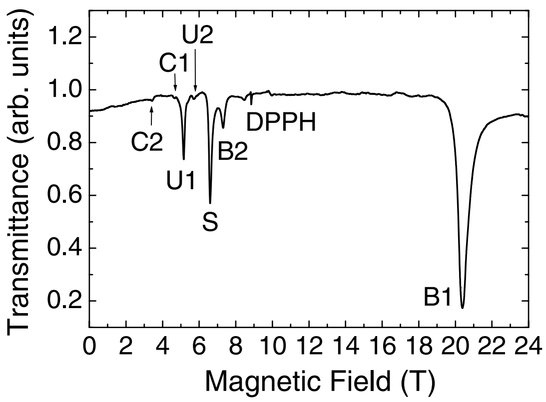

Figure 1. The ESR transmission spectra in Cu-PM, taken in the Voigt configuration at a frequency of 245.3 GHz at $T = 1.6$ K (for explanations see the text). DPPH was used as a marker.

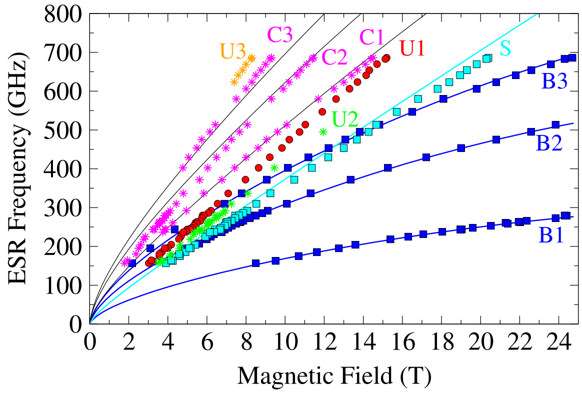

Figure 2. (Color online) The frequency-field dependence of the ESR modes in Cu-PM at $T = 1.6$ K. Symbols denote the experimental results, and lines correspond to contributions from specific excitations as predicted by the sine-Gordon theory (see the text for details).

Several resonance modes with different intensities were observed in the experiments. A typical ESR transmittance spectrum obtained in the Voigt configuration (with a propagation vector perpendicular to the external magnetic field) at a frequency of 245.3 GHz and at temperature $T = 1.6$ K is shown in Fig. 1. The mode B1 corresponds to the most intensive excitation, while the integrated intensities of the modes S, B2 and B3 are approximately six, twenty and two hundred times smaller, respectively. The complete frequency-field diagram of the observed magnetic excitations, collected both, in the Voigt and Faraday geometries, at $T = 1.6$ K, is presented in Fig. 2.

The experimental frequency-field diagram has been analyzed in the framework of the quantum sine-Gordon field-theory approach [2,3]. We used the following expression [3] for the soliton gap

Δ_s which is valid for a wide range of fields up to $g\mu_B H \sim J$:

$$\Delta_s = J \frac{2\Gamma(\frac{\xi}{2}) v_F}{\sqrt{\pi}\Gamma(\frac{1+\xi}{2})} \left[\frac{g\mu_B H}{J v_F} \frac{\pi \Gamma(\frac{1}{1+\xi}) c A_x}{2\Gamma(\frac{\xi}{1+\xi})} \right]^{\frac{1+\xi}{2}} . \qquad (1)$$

Here, c is the proportionality coefficient connecting the uniform applied field H and the effective staggered field $h = cH$, the parameter $\xi = (2/(\pi R^2) - 1)^{-1}$, where R is the so-called compactification radius, and v_F is the Fermi velocity. Both R and v_F are known exactly as functions of $\widetilde{H} = g\mu_B H/J$ from the solutions of the Bethe-ansatz equations [3]. The amplitude A_x, which is also a function of $\widetilde{H}$, was recently computed numerically [6]. At a given field H, there are $N = [1/\xi]$ breather branches B_n with $n = 1, \dots N$. The breather gaps Δ_n are given by the formula

$$\Delta_n = 2\Delta_s \sin(n\pi\xi/2). \qquad (2)$$

At $H = 0$ the first breather B_1 is degenerate with the soliton-antisoliton doublet S, $\overline{S}$. At finite H this degeneracy is lifted, so that B_1 becomes the lowest excitation and gives the strongest contribution into the magnitude of the gap observed in specific-heat experiments [7]. As follows from Ref. [9], the field-induced gap in $S = \frac{1}{2}$ antiferromagnetic chains with alternating g-tensor and/or the DM interaction is created by a soliton. This statement clearly contradicts our interpretation, where the gap is formed by the first breather. The sine-Gordon model predicts two more "heavy" breathers, B_2 and B_3, to exist in the relevant frequency-field range.

ESR probes the dynamical susceptibility $\chi(q, \omega)$ at the momentum $q = 0$. However, in the present case the alternation of the g-tensor and DM interaction leads to a mixing of $q = 0$ and $q = \pi$ components. Moreover, due to the fact that the Dzyaloshinskii vector is directed at an angle of about 58° with respect to the magnetic field [7], the physical susceptibility χ_{phys} is generally a mixture of effective longitudinal and transverse susceptibilities χ_{sG} calculated within the sine-Gordon model. As a result, one should be able to observe contributions from $\chi_{sG}^{\perp}$ as well as from $\chi_{sG}^{\parallel}$ both in the Faraday and Voigt geometries.

Single-particle contributions to the longitudinal susceptibility, $\chi_{sG}^{\parallel}$, are determined by solitons concentrated around incommensurate wave vectors $q = \pi \pm k_0$ and by breathers concentrated around $q = 0$. Here the incommensurate shift $k_0 = 2\pi m$ is determined by the total magnetization per spin m, exactly known as a function of field; for small H one has $J v_F k_0 \approx g\mu_B H$. In the transverse susceptibility, $\chi_{sG}^{\perp}$, the dominating contribution comes from breathers at $q = \pi$ and solitons at $q = \pm k_0$. Thus, in an ESR experiment there should be several breather resonances at the energies Δ_n as well as a single soliton resonance at

$$E_s \simeq \sqrt{\Delta_s^2 + (J v_F k_0)^2}. \qquad (3)$$

This exhausts the set of possible single-particle resonances; apart from them, there might be various multiparticle continua contributing to the spectrum (Fig. 2).

It is important to mention that in Eqs. (1)-(3) *the only* free fitting parameter is the coefficient c. Setting $c = 0.080 \pm 0.002$, we were able to achieve an excellent fit to the lowest observed mode B1, which is described by the first breather gap Δ_1. Using this value of c, we calculated the energies of other modes predicted by the sine-Gordon model and obtained a reasonably good fit to the entire set of the experimental data. On the basis of the fit, we identify the observed resonances as follows: the modes B1, B2, and B3 correspond to the first three breather resonances at Δ_1, Δ_2 and Δ_3, respectively (Fig. 2). The mode S is well fitted by the soliton resonance at E_s. This interpretation is supported by the analysis of the temperature dependence of the ESR spectra, which shows that the S mode continuously transforms into the $\omega = gH$ resonance when the temperature increases, in agreement with the theory [10]. The overall agreement between the sine-Gordon-theory predictions and the experimentally obtained frequency-field dependences for those four single-particle resonances is very good.

A detailed study of the temperature evolution of the ESR spectrum in Cu-PM in the perturbative spinon regime (*i.e.* when the temperature is high enough to destroy the soliton-breather superstructure, but sufficiently small compared to the characteristic energy of the exchange interaction J) is reported in [11]. It is important to mention that the staggered-field parameter c obtained from the analysis of the temperature dependence of the ESR linewidth and g-factor of magnetic excitations in the Cu-PM in the perturbative spinon regime ($c = 0.083 \pm 0.002$) is in excellent agreement with the value found by us from the analysis of the frequency-field dependence of magnetic excitations in the soliton-breather phase (Fig. 2).

The identification of the other six high-frequency ESR modes is more challenging since the theory does not predict any single-particle contributions in the relevant frequency region (for a more detailed discussion please refer to Ref. [12]). One may nevertheless speculate that the modes C1-C3 (Fig. 2) correspond to the edges of the soliton-breather continua. For the remaining three modes U1-U3 we were not able to find any appropriate explanation on the basis of the sine-Gordon model, although we checked all possible two- and three-particle continua. This is especially puzzling in case of the U1 mode, which is a very intensive excitation (see Fig. 1). Interestingly, an unexplained excitation, very similar to U1 in Cu-PM, was observed earlier in another $S = \frac{1}{2}$ sine-Gordon spin-chain material, copper benzoate [9].

In summary, we have presented a detailed frequency-field diagram of spin excitations in Cu-PM, a material containing $S = \frac{1}{2}$ AF chains with alternating g-tensor and DM interaction and exhibiting a field-induced gap. The use of high-resolution submillimeter-wave ESR spectroscopy made possible to obtain a very precise information on the magnetic excitation spectrum. The field-induced gap was observed *directly*, and its high-field behavior was studied. *Ten* ESR modes were resolved and their behavior in a broad field range up to $g\mu_B H \sim J$ was studied. By comparing the entire set of data with theoretical predictions, we have provided experimental evidence for a number of predicted excitations of the sine-Gordon theory, including *solitons* and the *three* lowest members of the breather hierarchy.

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