

Electron spin resonance in $S = \frac{1}{2}$ Heisenberg chains with alternating g -tensor and the Dzyaloshinskii–Moriya interaction [☆]

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Abstract

Low-temperature ESR studies of copper pyrimidine dinitrate, a spin- $\frac{1}{2}$ antiferromagnetic chain with alternating g -tensor and the Dzyaloshinskii–Moriya interaction, are presented. The ESR linewidth and g -factor shift are investigated as function of temperature. The data are interpreted in frame of a new theoretical concept proposed recently by Oshikawa and Affleck [Phys. Rev. Lett. 82 (1999) 5136] and applied for precise calculations of the ESR parameters in $S = \frac{1}{2}$ antiferromagnetic chains in the perturbative spinon regime. Excellent quantitative agreement between the theoretical predictions and experiment is obtained.

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Electron spin resonance (ESR) is traditionally recognized as one of the most powerful and sensitive tools for probing magnetic excitations in exchange-coupled spin systems. Using the temperature or magnetic field as a tuning parameter, one can obtain valuable information on the nature of the ground state and estimate their important physical parameters and constants. The low-dimensional spin systems are of particular interest for ESR. However, the lack of a proper ESR theory often made a detailed interpretation of experimental data and thus an accurate comparison with proposed models rather problematic. A new theoretical approach for calculating ESR parameters of $S = \frac{1}{2}$ AFM chains, which is based on bosonization and the standard Feynman–Dyson self-energy formalism, has

been recently developed by Oshikawa and Affleck (OA) [1]. The OA theory allows a *precise* calculation of the ESR parameters and their dependence on temperature and magnetic field. Importantly, the new concept avoids the Hartree–Fock approximation [2], which appears to be generally invalid for low-dimensional magnets. Here, we report a detailed ESR study of copper pyrimidine dinitrate ($[\text{PM-Cu}(\text{NO}_3)_2(\text{H}_2\text{O})_2]_n$, PM = pyrimidine; hereafter Cu–PM), a spin- $\frac{1}{2}$ antiferromagnetic chain with alternating g -tensor and the Dzyaloshinskii–Moriya (DM) interaction, which appears to be the most perfect realization of the quantum sine-Gordon spin-chain system known to date [3]. The measurements were performed in the intermediate (“perturbative spinon”) temperature regime, $E_g < T < J$ (where E_g is the size of the field-induced gap, created by the first breather excitation and J is the exchange coupling), at several frequencies, allowing us to check the temperature and field dependences of ESR parameters, predicted by the OA theory.

Cu–PM crystallizes in a monoclinic structure belonging to the space group $C2/c$ with four formula units per unit cell [3]. The Cu ions form chains running parallel to the

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short ac diagonal. The local principal axis of each octahedron is tilted from the ac plane by $\pm 29.4^\circ$. Since this axis almost coincides with the principal axis of the g -tensor, the g -tensors for neighboring Cu ions are staggered. In the presence of such structure, application of a uniform external field H induces an effective transverse staggered field $h = cH$, which leads to the opening of an energy gap $E_g \propto H^{2/3}$ observed in Cu-PM *directly* [4].

In general the Hamiltonian of a spin- $\frac{1}{2}$ Heisenberg AFM chain in an external field is given by

$$\mathcal{H} = J \sum_i \vec{S}_i \cdot \vec{S}_{i+1} + g\mu_B H \sum_i S_i^z + \mathcal{H}_\delta, \quad (1)$$

where the first term corresponds to the isotropic Heisenberg interaction, the second one is the Zeeman term and the third one represents various possible small anisotropic contributions. In accordance with the OA theory [1], the staggered field results in the symmetry-breaking perturbation \mathcal{H}_δ on the Green function with the self-energy contribution Σ whose real and imaginary part determine the ESR frequency shift and the linewidth, respectively. For the first time, the significant changes in the ESR spectra were revealed in the spin- $\frac{1}{2}$ chain systems copper benzoate [5] and $\text{BaCu}_2\text{Ge}_2\text{O}_7$ [6].

In our experiments high-quality Cu-PM single crystals were used, with the magnetic field applied along the c'' direction [3] providing a maximum staggered field. The ESR absorptions were fit using the Lorentzian formula for the line shape.

The temperature dependence of the linewidth has been studied at three frequencies, 9.4, 93.1, and 184 GHz. The corresponding data are summarized in Fig. 1, together with results of the OA theory. The temperature dependence of the effective g -factor in Cu-PM for a frequency of 184 GHz is presented in Fig. 2, together with calculations based on the OA theory [1] and using parameters obtained from the linewidth fit. We obtain a very good description of the *entire set* of the linewidth and g -factor shift data, as shown in Figs. 1 and 2, using $c = 0.083 \pm 0.001$. It should be noted that the inclusion of logarithmic corrections turns out to be important: neglecting them makes it impossible to achieve a *uniformly* good fit for the different frequencies. The obtained staggered-field parameter c is in excellent agreement with the value $c = 0.08 \pm 0.002$ found by us earlier from the analysis of the frequency-field dependence of ESR modes in Cu-PM in the soliton-breather regime [4]. More detailed studies of the field dependence of the ESR linewidth in Cu-PM can be found in Ref. [7].

In conclusion, we have presented a detailed study of the temperature evolution of the ESR spectrum in Cu-PM, a material which is considered to be the best available realization of a spin- $\frac{1}{2}$ Heisenberg chain system with alternating g -tensor and the DM interaction, in the perturbative spinon regime. The data were analyzed in frame of the OA quantum-field theory [1]. An excellent *quantitative* agreement with the theory has been achieved.

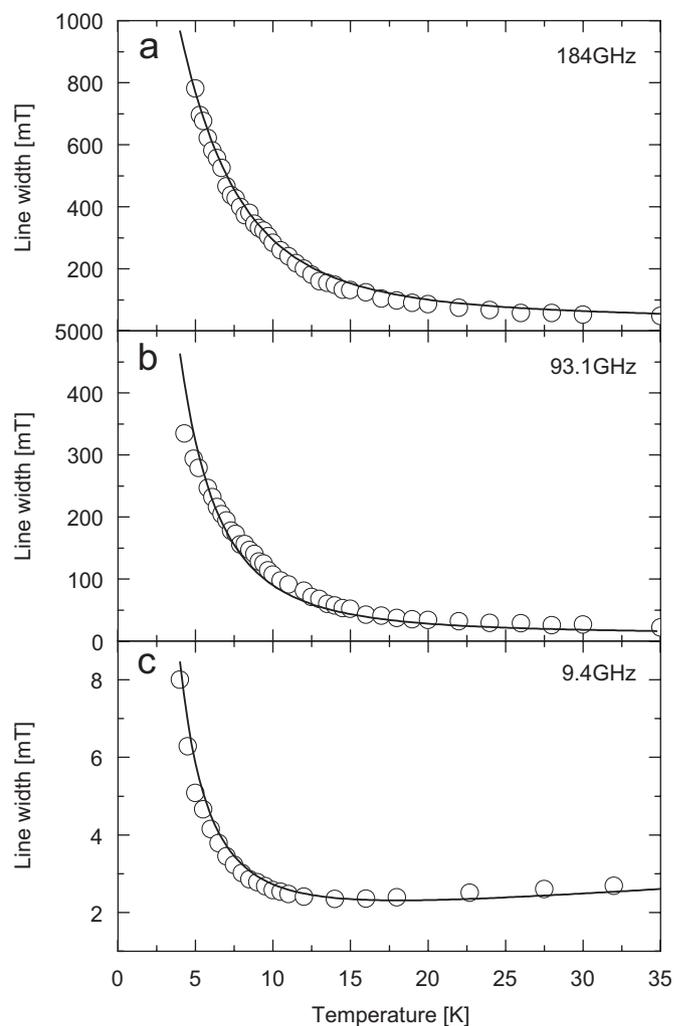


Fig. 1. The temperature dependence of the ESR linewidth in Cu-PM at frequencies of 184, 93.1, and 9.4 GHz. Symbols denote the experimental results, and solid lines correspond to the best fit using the OA theory [1].

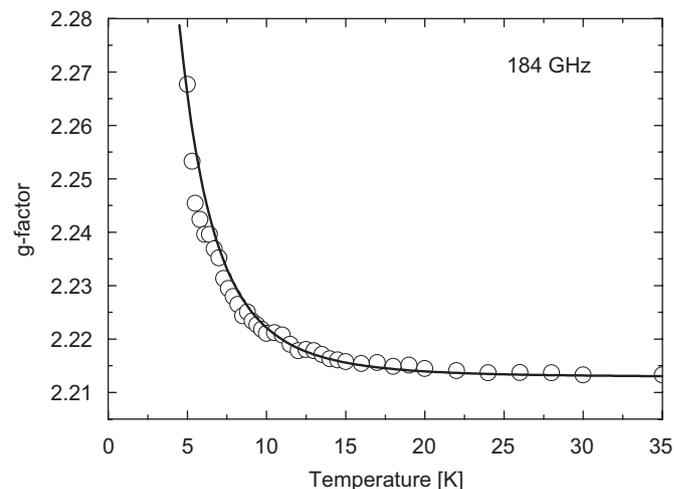


Fig. 2. The temperature dependence of the effective g -factor in Cu-PM for a frequency of 184 GHz. The solid line corresponds to calculations based on the OA theory [1] with the parameters obtained from the linewidth fit.

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