ESR Study of Sine-Gordon Excitations in S=1/2 Antiferromagnetic Chain: Copper Benzoate

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Abstract

We investigate electron spin resonance (ESR) on Cu benzoate, $Cu(C_6H_5COO)_2{\cdot}3H_2O$, which is an ideal model substance of S=1/2 antiferromagnetic Heisenberg chain (AFHC) with staggered field. Theoretically, S=1/2 AFHC subjected to the staggered field exemplifies a good realization of a quantum sine-Gordon (SG) model. In fact, under magnetic fields, this compound develops an energy gap $E_{\rm g}(H)$ in the low-lying SG excitation spectra as a function of the applied magnetic field H. In the previous work, we observed the SG-breather excitation for the first time, in wide-field range up to 30 T by means of ESR under Faraday configuration. In this work, we report a direct observation of the SG- soliton/antisoliton excitation by ESR under Voigt configuration. Thus, all solutions of the SG equation are now experimentally demonstrated by means of ESR technique.

Key words: Quantum spin; Field-induced gap; Cu benzoate; ESR

Recently, Dender *etal.* performed neutron scattering on deuterated Cu benzoate, $Cu(C_6D_5COO) cdot 3D_2O$ [1], the well-known S=1/2 antiferromagnetic Heisenberg quantum spin chain (AFHC) with an exchange coupling $J/k_{\rm B}$=8.6 K since 1960s [2]. Under magnetic fields, they observed the characteristic behavior of spinon excitations; new low-energy modes appear at the field-dependent incommensurate wave vectors in contrast with classical chains. However, the modes are not completely soft unexpected for S=1/2 AFHC system. Furthermore, the specific heat measurements under magnetic fields revealed an unusual energy gap as a function of external magnetic field; $E_{\rm g}(H) \propto H^{2/3}$.

As is explained by field theoretical approaches [3–6], the observed field-induced gap is caused by an effective transverse staggered field arising in proportional to the applied field, due to the staggered g-tensor and the Dzyaloshinskii-Moriya interaction. Theoretically, the S=1/2 AFHC system with the transverse staggered field is mapped to the well-known quantum sine-Gordon (SG) model. The spectra of the low-energy SG excitations consist of characteristic elementary excitations such a soliton, antisoliton and their bound state, which is called the breather.

Quite recently, we applied high-frequency and high-field electron spin resonance (ESR) measurement on Cu benzoate under Faraday configuration [7]. We successfully observed the breather excitation at low temperature and the field-dependent energy gap under a very wide field-range up to 30 T at 0.5 K. We also observed a certain quantum phase transition, which is a dynamical crossover between the gapless spinon regime and the gapped breather regime in the temperature versus magnetic field plane. Most recently, we endeavored to find a three dimensional magnetic long range order (LRO) by means of muon spin rotation/relaxation (μSR) measurement [8]. Under zero

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field, we couldn't observe any anomalies caused by the LRO down 20 mk by ZF-mSR. It is evident that Cu benzoate is the best candidate which exemplifies a good realization of the quantum SG model and it is of particular interest to note that the quantum critical behavior of S=1/2 AFHC can be tuned by the applied magnetic fields.

The main purpose of the present work is experimentally to demonstrate the soliton/antisoliton excitation followed by the SG model by ESR technique on Cu benzoate, that is, it is to explore the residual mathematical solution of the SG equation.

ESR measurements have been performed using a vector network analyzer in the frequency region between 53 and 109 GHz using a superconducting magnet and conventional Gunn oscillators and optical pumped fir-infrared laser combining pulsed magnetic fields for higher frequency region. All measurements were applied under Voigt configuration, where a propagation vector of the radiation is perpendicular to the external magnetic fields. That is to say, we can detect the parallel component of the fluctuations with respect to the applied magnetic fields. We have applied the magnetic fields parallel to the c-axis corresponding to one-dimensional chain. The c-axis is much more subjected to the contribution of the staggered fields than other axes [9].

Figure 1(a) shows ESR spectra for various frequencies for $H\|c$ at 0.5 K under Voigt configuration. The spectra at all frequencies have two parts of lines, one is a main line at higher field produced by the soliton/antisoliton excitation and the other at lower field has some kinds of lines expected to include very interesting and exciting complicated excitations; the higher order soliton/antisoliton excitation and the breather excitation corresponding to the perpendicular components of the fluctuations with respect to the applied magnetic fields. The detailed analysis of the frequency- and temperature-dependencies for the lines is discussed in separate paper [10]. The magnitude of the resonance field for main line corresponds to the energy gap caused by the soliton/antisoliton excitation, $h\nu$=$E_{\rm g}(H)$. The field dependent energy gap involving the results for the higher frequency data in the pulsed magnetic fields is shown in Fig. 1(b). The solid line is the theoretical curve of $E_{\rm g}(H)\propto H^{2/3}$ and the prefactor of this function is taken from one for breather excitation in Ref.[8]. The curve is well described with the field dependence of the energy gap below about 10 T. The obvious deviation at high-field region may arise from failure and breaking of the quantum SG model beyond applicability.

In conclusion, we have performed high-frequency and high-field ESR measurements on Cu benzoate under Voigt configuration. We have directly observed soliton/antisoliton excitation and nonlinear

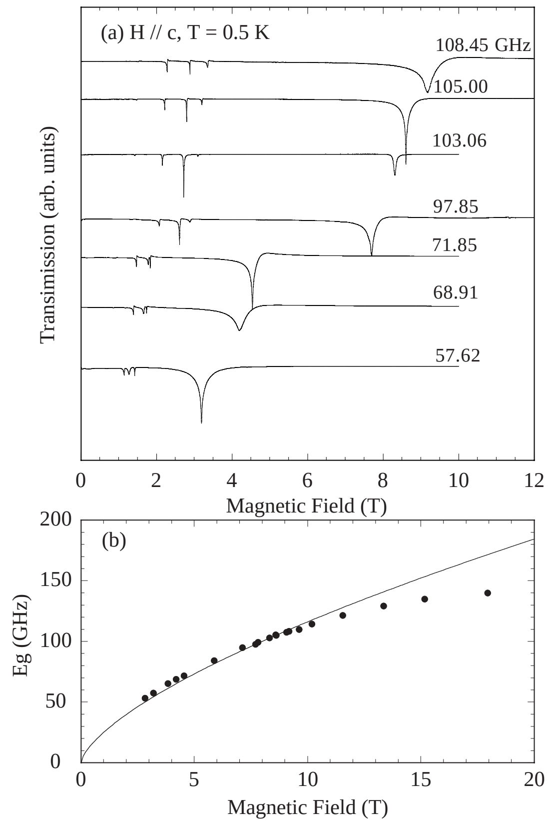

Fig. 1. (a) ESR spectra for various frequencies for $H\|c$ at 0.5 K. (b) Field dependence of the soliton/antisoliton energy gap $E_{\rm g}(H)$ for $H\|c$ at 0.5 K. Solid line is the theoretical curve of $E_{\rm g}(H)\propto H^{2/3}$.

$E_{\rm g}(H)\propto H^{2/3}$. The whole picture of the SG excitations for the S=1/2 AFHC with the staggered fields has been revealed experimentally by ESR technique.

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