Effect of Staggered Field in S=1/2 Antiferromagnetic Chain: Copper Pyrimidine

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Abstract

We report electron spin resonance (ESR) and magnetization measurements of the S=1/2 antiferromagnetic Heisenberg chain, Cu pyrimidine, $[\mathrm{PM{\cdot}Cu(NO_3)_2{\cdot}(H_2O)_2}]_n$(PM=pyrimidine). The effect of staggered fields due to both the alternating g-tensor and the Dzyaloshinskii-Moriya interaction is clearly observed for the ESR linewidth and magnetization.

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

An S=1/2 antiferromagnetic Heisenberg chain (AFHC) has received considerable attention for its remarkable quantum effect. Here, we are interested in the quantum AFHC system with the transverse staggered field, for which rich physics is involved in relation to the exactly solvable model of quantum sine-Gordon (SG) chain. Cu benzoate is the first example which represents the model system subjected to an effective transverse staggered field arising in proportion to the applied external field, due to the alternating g-tensor and Dzyaloshinskii-Moriya (DM) interaction. In good agreement with the theoretical prediction based on field theoretical approaches[1], it has been shown experimentally by neutron scattering, specific heat[2], and electron spin resonance (ESR) measurements[3] that Cu benzoate exhibits the field-induced energy gap in the SG excitation spectrum as a function of external magnetic fields, $E_\mathrm{g}(H)\propto H^{2/3}$. It is particularly important to provide other examples of real substances in order to inquire whether the observed behaviors are specific only to Cu benzoate or generic to the quantum system with the transverse staggered field.

Quite recently, Feyerherm *et al.*[4] have discovered another candidate, Cu pyrimidine, $[\mathrm{PM{\cdot}Cu(NO_3)_2{\cdot}(H_2O)_2}]_n$ (PM=pyrimidine) with J/k_B=36 K which has the crystallographically feature very similar to that of Cu benzoate. Indeed, they revealed the angle-dependent field-induced gap as a function of the external magnetic field by specific heat measurements and determined that the largest staggered field and, hence, the largest energy gap are induced for the field direction along the c''-axis in the ac-plane from the analysis of angular dependent-susceptibility. The main purpose of the present work is to clarify experimentally the effect of the transverse staggered field on the ESR linewidth and magnetization of Cu pyrimidine.

Figure 1(a) shows the temperature dependence of linewidth ΔH_pp, defined as a peak-to-peak of ESR field-derivative signal at X-band. It is remarkable that the temperature dependence is drastically different between two field directions parallel and perpendicular to the c''-axis in the ac-plane. For $H\|c''$, the linewidth shows a divergent broadening proportional to T^{-2} at

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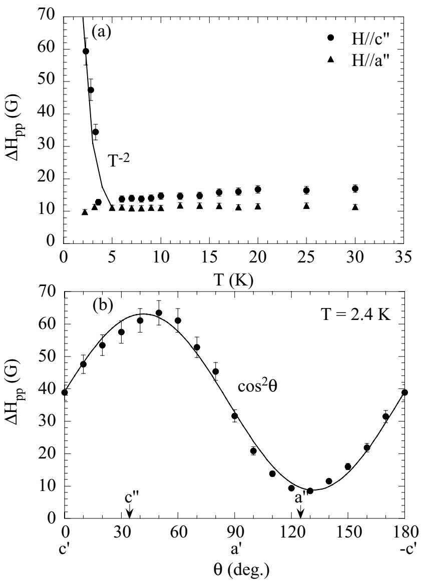

Fig. 1. (a)Temperature dependence of linewidth for $H \| c''$ and $H \| a''$. (b)Angular dependence of linewidth at 2.4 K in ac-plane on Cu pyrimidine

lower temperatures. In contrast, for $H \| a''$, which is perpendicular to the c''-axis and corresponds to the direction which we expect to be practically free from the effect of staggered field, the linewidth shows no broadening with decreasing temperature. A divergent broadening is often observed as the temperature approaches to the three-dimensional ordering. However, the present case is highly unusual in view of that there is a specific direction for which we observe no broadening. As shown in Fig. 1(b), the linewidth at the lowest temperature of 2.4 K changes quite systematically as a function of the field direction in the ac-plane and the angular dependence of the divergent contribution is well described by $\cos^2\theta$-law. According to the theory by Oshikawa and Affleck[1], the linewidth for the AFHC with the staggered field is expected to show a divergent broadening as $\Delta H_{\rm pp} \propto (h/T)^2$ in the temperature range $T \ll J/k_{\rm B}$ where h is the effective staggered field. This prediction is in good agreement with our observation both in the temperature dependence and the angular dependence of the divergent contribution because h is naturally expected to be proportional to $H\cos\theta$. It is interesting to note that almost the same angular dependence is observed for the divergent contribution of magnetic susceptibility due to the effect of the staggered field as reported in Ref.[4]. Also, it is interesting to note that the c''-axis does not coincide with any principal axes of alternating g-tensor, indicating the coexistence of the DM-interaction. The detailed analysis for the temperature and angular dependencies of the linewidth is discussed in Ref.[5].

Figure 2 shows the magnetization process at 1.3 K

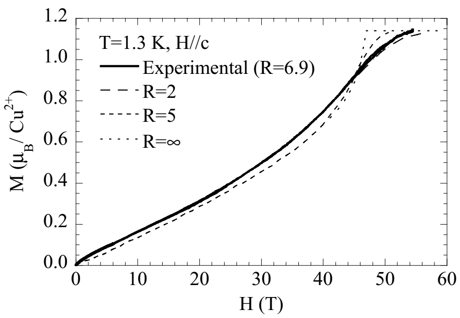

Fig. 2. Magnetization process of Cu pyrimidine for $H \| c$ at 1.3 K.

for $H \| c$ up to 54 T. It is estimated that the saturation field is about 47 T(=$2J/g\mu_{\rm B}$) using $J/k_{\rm B}$=36 K and g=2.28. The dashed lines are the theoretical curves for AFHC with no staggered field at zero and finite temperatures by Inawashiro and Katsura[6] and do not agree with the experimental data. The most striking difference is that the experimental curve show a monotonic change and no singular behavior at the saturation field. It is evident that this is not due to the effect of finite temperature as seen from the comparison with the calculation and, more confidently, from the experimental fact that the data at 4.2 K is almost the same though the R=$J/4k_{\rm B}T$ factor increases three times at 1.3 K. It is natural to ascribe this discrepancy to the effect of staggered field since the sharp transition at the saturation field is expected to be obscured in the presence of conjugate field to the antiferromagnetic staggered magnetization. In fact, a recent calculation for AFHC with staggered field[7] explains no singular behavior at the saturation field as well as an initial sharp increment near zero field in agreement with our observation.

In conclusion, we have clearly observed the effect of the staggered fields due to both the alternating g-tensor and the Dzyaloshinskii-Moriya interaction on the ESR linewidth and magnetization of the recently discovered compound, Cu pyrimidine.

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