ESR Study of Frustrared Δ-Chain System

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Abstract

High field ESR measurements were performed in Δ-chain compound $[Cu(bpy)H_2O][Cu(bpy)(mal)H_2O](ClO_4)_2$, which consists of spin trimers with ferromagnetic and antiferromagnetic competing exchange interactions. A narrow EPR absorption line of Cu^{2+} and its anomalous temperture depndence were observed. The g-shift and frequency-dependent linewidth of this substance in the low temperature region are discussed.

Key words: ESR ; One-dimensional system ; Dynamical bahavior ; linewidth ; g-shift

Quantum spin chains have been studied externsively both experimentally and theoretically. Due to the development of short range order at low temperature, the dynamical properties of one-dimensional (1D) system appear in the peculiar temperature dependence of the linewidth and g-shift in many compounds, which can be well reproduced by the ESR theory for 1D antiferromagnet (AFM) by Nagata and Tazuke (NT) [1]. However it is well known that some Cu compounds cannot be fully explained by this theory[2]. Recently, Oshikawa and Affleck (OA) proposed a new ESR theory using the field theoretical approach[3] and this theory showed good correspondences with the temperature dependences of the linewidth and resonance shift of the Cu benzoate[4]. In this paper, we studied new S=1/2 1D spin system called "Δ-chain". The Δ-chain forms a saw-tooth lattice consisting of frustrated S=1/2 spin trimers. The model substance of Δ-chain compound $[Cu(bpy)H_2O][Cu(bpy)(mal)H_2O](ClO_4)_2$ was synthesized by Ruiz-Pérez *et al* [5]. There are two kinds of nearest-neighbor exchange interactions, J_1 between the basal spins is antiferromagnetic (AF) (-6 K), while J_2 between the apical and the basal spins are ferromagnetic (F) (6.6 K). To investigate the dynamical behavior of this Δ-chain compound, millimeter and submillimeter wave ESR have been performed.

ESR measurements have been performed using pulsed magnetic fields up to 30 T and frequency between 60-315 GHz[6–8]. A Bruker ESR spectrometer was used for 9.5 GHz. Single crystals were synthesized by one of the author (H. K.) and were analyzed by X-ray diffraction at Institute for Chemical Research of Kyoto University.

Figure 1 shows the temperature dependence of the g-values of EPR observed at 160 GHz and 9.5 GHz. The external field was applied to three kinds of directions as $B \parallel b$ (chain), $B \parallel c$, and $B \parallel x$. The x-direction lies in the a-c plane and is perpendicular to the b and c axis. The typical g-shifts in 1D AFM are found for $B \parallel b$ (positive) and $B \parallel x$ (negative), while g-value for $B \parallel c$ shows a positive shift . The positive g-shift observed for the unique direction, which is perpendicular to the chain, cannot be easily interpreted by NT theory or OA theory. From the crystallographic viewpoint, there should be inequivalent g-tensor sites in the basal spin

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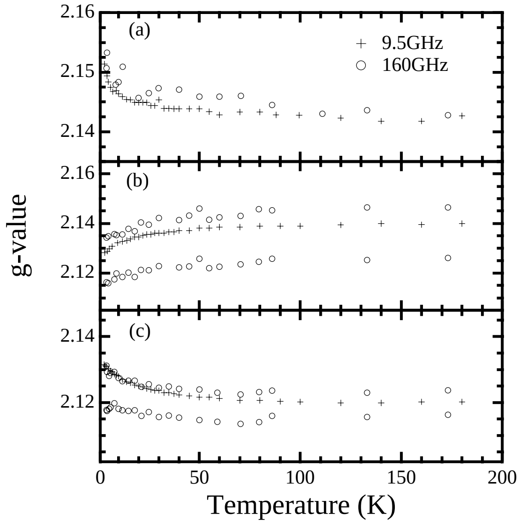

Fig. 1. Temeprature dependence of g-values observed at 9.5 GHz (+) and 160 GHz (○). (a) $B \parallel b$, (b) $B \parallel x$, (c) $B \parallel c$. The split of the lines for $B \parallel c$ and $B \parallel x$ is due to the bicrystal.

chains with AF interaction. This fact suggests that the g-values increase for all directions in the very low temperature region[3]. However, the exchange interaction due to the apical spins in the Δ-chain should affect the dynamical behavior of EPR, and we observe consequently anomalous g-shift in our Δ-chain compound.

Temperature dependences of the linewidth oberved at various frequencies are shown in Fig. 2. There is a clear field-dependent (in other words frequency-dependent) diverging behavior in the low temperature region. From the crystallographic view point, it can be expected that the Dzyaloshinskii-Moriya (DM) interaction exists in this compond. The exsistence of staggerd field due to inequivalent g-tensor and DM interaction produces the strong field dependent power law behavior $(B/T)^2$ of linewidth[3,4]. For $B \parallel b$, we can observe the B^2 dependence of linewidth at 4.2 K, while for other directions, this kind of field dependences were not clearly observed. This field-direction dependence is in agreement with OA theory. However, the temperature dependence is not reproduced by T^{-2}. The discrepancy may be caused by the Δ-chain structure which cannot be described by a uniform 1D AFM with transverse staggerd fields. There is a possibility that the exchange interaction due to the apical spins affects the temperature dependence of EPR linewidth as same as g-value. This point remains as a future problem.

In summary, high field ESR measurements were performed in the Δ-chain compound, and anomalous g-shift and clear frequency-dependence of linewidth in the low temperature region are observed.

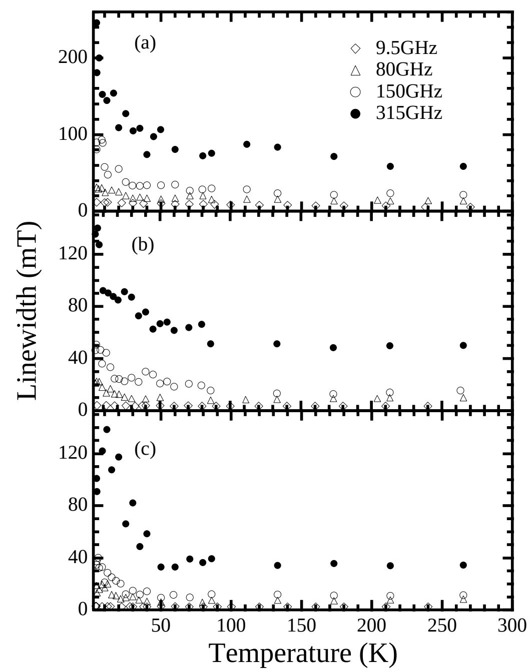

Fig. 2. Temeprature dependence of linewidth observed at 315 GHz, (●) 160 GHz (○), 80 GHz (△) and 9.5 GHz (◇), respectively. (a) $B \parallel b$, (b) $B \parallel x$, (c) $B \parallel c$.

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