Coexistence of gap-less and gapped excitations in NH_4CuCl_3

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Abstract

A low temperature ESR investigation is performed on a low dimensional compound NH_4CuCl_3 with magnetization plateaux. In the phase below the $\frac{1}{4}$–plateau, a coexistence of gapped two triplets and a gap-less mode is found. Below 1.3 K at which a part of spins show some magnetic ordering, the higher energy triplet mode exhibit no change. In addition to the previously reported gapped modes at $\frac{1}{4}$– and $\frac{3}{4}$–plateaux, a new low energy mode is found at the half of the saturation field. This mode may be caused by an "irrelevant instability" of $\frac{1}{2}$–plateau.

Key words: high magnetic field; spin gap; magnetization plateau; electron spin resonance

1. Introduction

A problem of a magnetization plateau has attracted considerable attentions in this decade. Theoretically, the necessary condition for the appearance of a plateau was given by Oshikawa, Yamanaka and Affleck(OYA)[1] in case of a spin chain with axial symmetry. A dimer system NH_4CuCl_3 has been known as one of model substances of this subject. In the compound, the $\frac{1}{4}$– and $\frac{3}{4}$–plateaux are observed and excitation gaps are also found at those plateaux. [2,3] Since a gradual increase of magnetization is suppressed by an excitation gap, the appearance of gap and the observation of the plateau are consistent each other. For this typical behavior, NH_4CuCl_3 has been considered as a good "realization" of magnetization plateau. While such ideal behaviors are found , several anomalies are also observed. First of all, the absence of $\frac{1}{2}$–plateau is mysterious because it is generally beleaved that a $\frac{1}{2}$–plateau is more robust than a $\frac{1}{4}$–plateau. Another anomaly is that the system undergoes three-dimensional(3D) magnetic ordering at $T_N = 1.3$ K.[4] From the specific heat, it is estimated that one-quarter of the spins are in the ordered state and the remaining spins are in the singlet state. Such coexistence of the gapped state and the 3D magnetic ordering is very unusual. The nature of the ground state has not been investigated microscopically and thus no model has been proposed for the anomalous phase transition. Since the previous ESR measurement was performed at 1.5 K, we extend, in the present work, the temperature range down to 0.5 K to investigate the low temperature phase below 1.3 K.

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2. Results and discussions

ESR experiments have been performed by using two different high frequency ESR systems.[5,6] The measurements have been performed below 2 K and hence, excitations from the ground state to the excited states and excitations within the ground state are observed. Transitions among the excited states are not observed at this temperature range.

Below the first plateau appearing between 4.7 and 13.3 T, we observed two excited triplets. The excita-

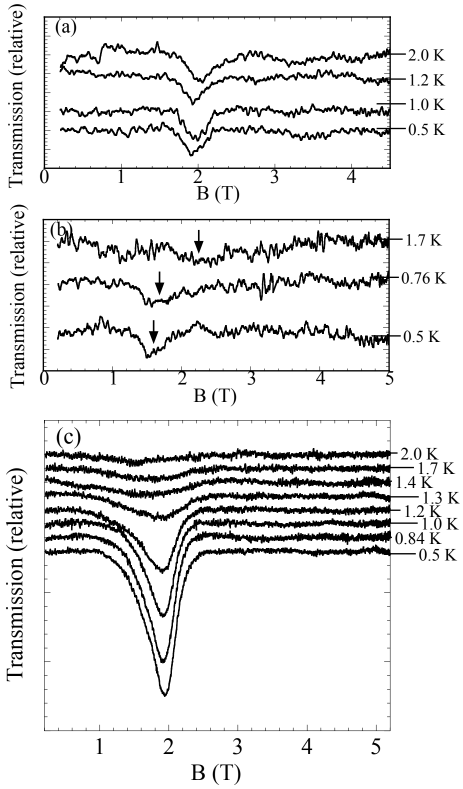

Fig. 1. Temperature dependence of ESR spectrum for $H \| a$ at (a)761.6 GHz, (b)430.9 GHz and (c)323.7 GHz

tion gaps at zero field are 360 GHz and 710 GHz. Temperature dependence of the ESR signal of the higher energy triplet is shown in Fig. 1(a). No temperature variation of the spectrum is found between 2 K and 0.5 K. It indicates that this triplet is not influenced by the 3D ordering at 1.3 K. This observation is an experimental proof of the coexistence of a singlet state and a magnetic ordering. The temperature dependence of the lower energy triplet is more complicated as shown in Figs. 1(b) and 1 (c). The up-going branch of the triplet at 430.9 GHz shows a small enhancement of the intensity. On the other hand, a considerable increase of the intensity is found for the down-going branch at 323.7 GHz. The onset of the observed anomaly is 1.3 K and thus the change should be related to the 3D ordering. While an anomaly is found for the intensity, no large shifts of the resonance fields are found for these two branches. This fact clearly shows that the low energy spin gap also survives under the 3D ordering. The down-going branch is related to a spin parallel to the external magnetic field. Hence, the increase of the intensity may be caused by the enhancement of the spin polarization of this branch which is originated from the 3D ordering.

The signal of the spins involved in the 3D ordering is also observed as a straight extension of the paramagentic ESR signal from high temperature. In a spin gap compound, the intensity of a paramagentic signal decreases rapidly where the temperature is lower than the energy of a spin gap. In NH_4CuCl_3, a strong intensity of the paramagentic signal is found even at 1.5 K. Since the temperature is much lower than the spin gap, it is considered that the signal is caused by a part of spins in the gap-less state. If the transition at 1.3 K is an antiferromagentic ordering, antiferromagnetic resonance modes with anisotropy gaps should present. No clear change of this paramagentic signal is found at the transition. It suggests that a 3D ordering found by the specific heat experiment is not a convectional antiferromagentic ordering. Although the coexistence of a gapped state and a gap-less state is clearly shown by ESR, no microscopic model for the low temperature phase is possible at present.

Finally, we mention a new gapped mode found at the half of the saturation field. We found a ESR absorption at 15.5 T below 50 GHz. Since the magnetization is about half of saturation magnetization at this field, it is natural to attribute this mode to the anomaly induced by a hidden $\frac{1}{2}$–plateau. It is evident that the low energy mode is gapped and thus the system has a small excitation gap at $\frac{1}{2}$–magnetization. The missing of a real plateau may be interpreted by the disturbance by the gap-less excitation. Future theoretical investigations are also needed for this point.

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