Electron Spin Resonance Measurements at Ultralow Temperatures

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Abstract

An electron spin resonance (ESR) measurement system for use at ultralow temperatures using a ^{3}He-^{4}He dilution refrigerator has been developed down to 160 mK. As the first experiment, the ESR was measured on a quantum spin chain Cu benzoate, in which a field-induced gap was recently found. The evaluation of a new ESR mode, so called breather mode, was found at the lowest temperatures. In the present work, we observed a smaller gap in $H\|$b by suppressing of the thermal excitation.

Key words: Electron Spin Resonance; Dilution Refrigerator; Quantum Spin Chain; Cu-Benzoate

1. Introduction

Magnetic resonance is effective for observing the dynamical physical properties in a spin system with quantum fluctuations. NMR and μSR measurements have been carried out at ultralow temperatures and many notable results were achieved. In contrast, for ESR measurement, few experiments using a dilution refrigerator have been performed. This is because of technical difficulties such as heat leakage through the light guide and heating by microwave. In this paper, the development of an ESR measurement system for use at ultra low temperatures by using a vector network analyzer, which has ultrahigh sensitivity, is reported.

2. Experimental Settings

Figure 1 shows a schematic picture of the ESR system. ESR measurements have been performed using a vector network analyzer (AB millim$'$etre Co., Ltd.) in the frequency region between 54 GHz and 111 GHz using a 20 T superconducting magnet. Using the vector network analyzer, the full tunability of frequency enabled us to purge a mechanical tuner to adjust the length of a cavity, which is very difficult to do at ultralow temperatures. High-sensitivity measurements are possible using a cylindrical resonant cavity. In order to achieve a good thermal contact between the sample and the cavity, the sample was fixed directly on the endplate of the cavity. The cavity is installed in a ^{3}He-^{4}He dilution refrigerator (Kelvinox System, Oxford Co., Ltd.). The cooling power is 25 μW at 100 mK. The cavity holder was fixed directly on the Cu plate, which is connected to the mixing chamber by a copper pipe covered with a stainless-steel pipe. Between the cavity and the analyzer, rectangular wave guides WR 22 made from Cu-Ni alloy of low thermal conductivity were used for the microwave transmission. In order to suppress the heat leakage from the room-temperature area, several heat anchors were attached at the still heat exchanger and the mixing chamber. Moreover, a sapphire plate and black pa-

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Fig. 1. ESR measument system using dilution refrigerator

per were inserted as a cold filter, as shown in Fig. 1. In order to cool the sample more efficiently, meshed Cu wires were connected between the cavity and the mixing chamber. The temperatures were measured by a ruthenium oxide thermometer (Scientific Instruments Co. Ltd.) attached to the cavity. The lowest temperature of the cavity was 155 mK.

The power of the microwave in the cavity is estimated to be less than one μW, which is much smaller than the cooling power of the dilution refrigerator. The heating of the sample was checked by varying the input power by more than 40 dB. The heating due to the microwave was only 2 or 3 mK at the lowest temperature. When the ESR measurement was carried out, the field sweep speed was very slow, about 0.05 T/ min, in order to avoid the Eddy current heating of the cavity. All measurements were performed in the Faraday configuration, where a propagation vector of the radiation is parallel to the external magnetic fields. Single crystals grown by the diffusion method and rectangular-shaped samples with typical dimensions of $0.5 \times 0.5 \times 0.1$ mm^3 were used for the measurements.

3. ESR study on one dimentional Quantum Spin Chain, Cu-Benzoate

As the first measurement, we studied Cu benzoate, which is an ideal one-dimensional quantum spin chain. It was discovered by Date et al. at 1960s and many studies on it have been carried out [1] [2]. In addition to the paramagnetic resonance, they observed a new low- temperature peak. It was proposed that the gap is caused by the staggered field originating from the alternatering g-tensors. This proposal was extended and

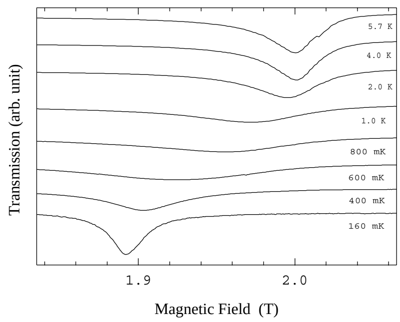

Fig. 2. ESR signal of Cu-Benzoate. The frequency is 57.423 GHz.

proved by the theoretical studies of Oshikawa and Affleck (OA), [3] [4] [5]Essler and Tsverik. [6] It is proposed that the staggered field is mainly caused by the alternative Dzyaloshinsky-Moriya vectors, and that, instead of spinon excitation, soliton, antisoliton and their bond state, a so called breather dominates the excitation of the spectrum. In our previous paper, we have performed precise ESR investigations of the $H \|$c-axis, which is the direction in which the field-induced gap is largest down to 0.5 K, [7] and found a clear ESR anomaly by a field induced gap. In the present paper, the achievement of ultralow temperature enabled us to study the behavior of the field-induced gap even in low-field region. We performed ESR study on Cu benzoate. By cooling the sample to ultralow temperatures, a well-defined breather excitation was observed for $H \|$b, the gap is smaller than that for $H \|$c-axis. The field dependence of the energy gap of the breather excitation agrees well with the results of the specific heat measurements. [7] We have also clearly observed the dynamical crossover between the spinon regime and the gapped breather regime. The authors would like to thank M. Yoshida for supporting our experiment. This investigation was performed at the High Field Laboratory for Superconducting Materials, Tohoku University. This work was supported by a Grant-in-Aid for Scientific Research from the Ministry of Education, Culture, Sports, Science and Technology of Japan.

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