Multiferrocity in the Gapless Quantum Antiferromagnet NH₄CuCl₃

We have developed a ESR and a magnetization probes in which a high electric field can be applied in addition to a high magnetic field. The highest voltage is about 1000 V at 4.2 K. It was enabled by the use of multiple thin polyimide tubes for electrode cover. At low temperatures, the voltage effect is not obvious, presumably because the barrier is too large to switch the polarization in the electric field of 1000 V.

Multiferroics compounds that can simultaneously exhibit long range magnetic and dielectric orders, have been the subject of intense study over the past decade. One reason is their great potential for a variety of practical applications, which includes data storage devices, data encryption, microwave devices, ac field sensors, transducers, actuators and more. Unfortunately, a number of fundamental physical restraints has resulted in a paucity of such materials, requiring a different approach to their synthesis and discovery.

One of the important question is the coupling between the magnetic and the electric orders. Also the coupling between the responses to the magnetic and the electric fields. The purpose of this project is to establish the experimental methods to measure the magnetization and the ESR under the coexisting of two kinds of fields-magnetic and electric fields.

As the target for this research, a new class of multiferroics, NH₄CuCl₃ is chosen. It is the three dimensionally coupled S=1/2 spin dimer system. The zero field ground state is gap-less and there are 1/4 and 3/4 magnetization plateau for the macroscopic quantization of the magnetization. For this compound, dielectric, specific heat, mass spectroscopy and X-ray measurements, ESR and other properties have been investigated. To date, there is no investigation of ESR in the presence of the electric field.

Figure 1 shows the schematic drawing of the sample mount in ESR probe in electric field. The sample is mounted on the polyethylene sample case with the bottom and top electrodes. The bottom one is connected to the light pipe made of SUS304 and then to the electric ground. The top one is connected to the high voltage electrode from the high voltage power supply. The sample is sealed into the stainless pipe with He-exchange gas. In the diluted He gas, the discharge takes place in the relatively low



Fig. 1 Schematic diagram of ESR setup.

voltage threshold of several hundreds volt. On the other hand, the thick cover on the high voltage line results in the high loss in the transmission of the electromagnetic wave. In the present system, we have used an ultra-thin polyimide tubes in the range of 0.1-0.6 mm in diameter. By putting those tubes with sufficient overlap, we could apply the high voltage as high as 1000 V to the sample.

We have measured at several frequencies to observe different ESR modes. At 190 GHz, we can observe the ground state ESR signal and the excitation from the singlet to triplet states. By allying the high voltage keeping the sample at 1.7 K, we see a small change of the base line, but there is no large change in the ESR spectrum. It may show the large barrier to modify the ferroelectric order in low temperatures.

We have also succeeded in applying the high voltage in the magnetization measurement specimen. However, the magnetization signal was weak for the used small sample with the diameter of 0.2 mm. A sufficient signal can be obtained by using the larger sample.

In summary, we have succeeded in the application of high voltage in the high frequency ESR and high field magnetization. The further investigation of the magnetic properties of NH₄CuCl₃ is in progress.

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