Dielectric Study of a New Phase Transition in an Organic Triangular Antiferromagnet TNN in High Magnetic Fields

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Introduction

S=1/2 spin trimers in which three spins are antiferromagnetically coupled with equal strengths are of strong current interest. When such spin units form a triangular lattice, theory predicts novel multiferroic effects which arise from mechanisms that are completely lost when the fundamental many-body hamiltonian (or a more simple Hubbard hamiltonian, for that matter) for the trimers is projected onto the spin sector to obtain a spin Hamiltonian [1]. However, experimental studies of such a system have been hampered by the lack of an ideal material. For instance, well known spin trimers, such as {Cu3}, suffer from a Jahn-Teller distortion [2]. As a result, they are predicted to exhibit only conventional multiferroic effects arising from a trivial Dzyaloshinskii-Moriya term in the spin Hamiltonian [3]. To overcome this limitation, our approach has been to synthesize crystals of organic triradicals that possess perfect three-fold-rotation symmetry. In our previous experiments, we have measured the specific heat of such a material, dubbed TNN, in magnetic fields between 1.3 T and 8.5 T, where no magnetic ordering occurs, in order to look for evidence for a ferroelectric transition predicted by the theory [4].

Experimental

In this magnetic field range, the specific heat was measured in a relaxation calorimeter developed by Professor Takano's group at the University of Florida. This instrument allowed measurements from 0.1 K to 5 K, without compromising the operation of the dilution refrigerator. We measured the dielectric constant by creating gold electrodes on the sample surface by sputtering and by measuring the capacitance between them with a commercial capacitance bridge operated at 5 kHz. We also attempted to detect the electric polarization of the sample by measuring pyroelectric current with an electrometer. The polling electric field used to polarize the sample was created by a dc voltage up to 400 V applied between the electrodes.

Results and Discussion

The theoretical expectation prior to the experiment was that the ferroelectric ordering may take place at a temperature too low for relaxation calorimetry. We were therefore prepared to detect any non-phononic specific heat at temperatures below about 0.2 K, indicative of the lifting of the twofold spin degeneracy—per trimer—of the ground state. Surprisingly, however, a sharp specific-heat peak due to a phase transition was found at a considerably higher temperature, 0.4 K, which is nearly field independent. Further theoretical work is needed to explain why the transition occurs at such a temperature, as high as the strength of the intertrimer exchange in TNN.In the measurements of the dielectric constant, a broad peak was detected roughly at a temperature where the specific-heat peak was found. However, no pyroelectric current indicative of an electric polarization was detected.

Conclusions

We have succeeded in the dielectric measurements of an organic spin trimmers and observed the clue of the novel multiferroic effects. To examine whether the dielectric-constant peak signifies a ferroelectric transition, we must make further studies of frequency dependence and pyroelectric-current measurements with an improved sensitivity.

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