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2007 J. Phys.: Condens. Matter 19 145237

(http://iopscience.iop.org/0953-8984/19/14/145237)

J. Phys.: Condens. Matter **19** (2007) 145237 (5pp) doi:10.1088/0953-8984/19/14/145237

Commensurate and incommensurate phases of the distorted triangular antiferromagnet Cs_2CuBr_4 studied using ^{133}Cs NMR

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Received 25 August 2006
Published 23 March 2007
Online at stacks.iop.org/JPhysCM/19/145237

Abstract
A quasi-two dimensional antiferromagnet Cs_2CuBr_4 is characterized as a frustrated spin system on a distorted triangular lattice with easy-plane type anisotropy. ^{133}Cs NMR experiments on Cs_2CuBr_4 have been performed in the range of magnetic field up to 15.9 T, applied along the b-axis, which covers the field range for the 1/3 magnetization plateau, in order to clarify the properties of the field-induced incommensurate–commensurate transition at both ends of the plateau. We found that the NMR spectrum shows hysteresis and two phase coexistence around the transition field. We also present a field dependence of T_1^{-1} at 0.5 K showing a hysteresis around each end of the plateau, indicating a first order incommensurate–commensurate phase transition.

1. Introduction

The triangular lattice antiferromagnet (TAF) is well known as a regular lattice with geometrical frustration. In particular, the spin-1/2 TAF presents a good example for investigating a quantum effect incorporated with a frustration effect. Furthermore, theoretical studies have revealed that a 1/3 magnetization plateau appears for a spin-1/2 TAF due to a quantum effect [1–3]. The presence of such a plateau was experimentally found in a spin-1/2 distorted TAF, Cs_2CuBr_4 [4]. In addition, the 2/3 plateau was found quite recently [5]. These plateaux are observed clearly in the magnetization curves in fields applied along both b- and c-axes. Thus, these plateaux are considered to be stabilized by the quantum fluctuation of a spin-1/2 TAF. In the crystal of Cs_2CuBr_4, $CuBr_4^{2-}$ tetrahedra constitute a chain along the b-axis due to the exchange interaction J_1, and there also exists another coupling J_2 between the chains. As a result, Cu^{2+} ions ($S = 1/2$) are arranged on an isosceles triangle-based lattice formed by J_1 and J_2 in the bc-plane. The coupling between the triangular lattice planes is considered to be very weak ($\sim 0.05 J_1$), by analogy with an isostructural compound Cs_2CuCl_4 [6]. Ono and co-workers estimated from the neutron diffraction measurements [5] the ratio $J_2/J_1 = 0.74$ which is rather close to unity as compared with that for Cs_2CuCl_4; when J_2/J_1 is unity, the system is characterized as an equilateral triangular magnet. They have also suggested a cycloidal incommensurate spin ordering and a commensurate spin ordering for outside and inside of the plateau region, respectively, below $T_N \simeq 1.4$ K. Therefore, when the field is increased below T_N, there are incommensurate–commensurate transitions at both ends of the plateau. It is interesting that both the long-range ordering and the magnetization plateau occur simultaneously.

In order to study the spin dynamics and the spin structure in Cs_2CuBr_4 from a microscopic viewpoint, we have performed ^{133}Cs NMR experiments in a temperature range down to 0.4 K and in the range of magnetic field up to 15.9 T applied along b-axis which covers the 1/3 magnetization plateau region (approximately 13.9–15.0 T for our sample).

2. Experimental method

We used the same sample of a single crystal Cs_2CuBr_4 throughout our measurements, because the critical fields of the 1/3 plateau slightly depend on the sample [7]. A conventional pulsed NMR method is used and the ^{133}Cs NMR spectrum was obtained as a function of the frequency under a constant magnetic field. As shown in our previous paper [8], two groups of NMR lines were observed in all cases, which is consistent with two inequivalent caesium sites, called A and B sites, in the crystal of Cs_2CuBr_4. In this paper, we devote consideration to the results for the B site.

In order to clarify the properties of a field-induced phase transition, it is important to perform experiments with carefully controlled external field. The field-induced transition was studied by the following procedure: for the measurement of increasing field, the applied field was increased monotonically step by step from a sufficiently low field; then, the data for decreasing field were obtained in the opposite way. In these experiments, the value of the magnetic field was checked by means of the resonance frequency of ^{63}Cu in the wire of NMR coil.

3. Experimental results and discussion

Figure 1 shows NMR spectra of the B site of ^{133}Cs measured at 0.45–0.46 K under an external magnetic field applied along the b-axis. The spectrum for the B site consists of a single line in the paramagnetic phase and splits into two peaks below T_N. At 13.5 T below the plateau region, the spectrum shows a double horn type line shape with a continuum of finite intensity between the two peaks. Such a line shape is known as a signature of an incommensurate spin structure. Generally, an NMR spectrum reflects the distribution of the component parallel to the external field of the hyperfine field caused at nuclei. In the cycloidal phase, the hyperfine field caused at caesium nuclei should have a continuous distribution. On the other hand, under a field above about 13.9 T, the continuum between the peaks disappears and the spectrum consists of two discrete peaks for each site. This is a feature of the commensurate phase and was observed at any position in the plateau region. Further, the ratio of the intensity of the two lines is 1:2, which suggests the realization of up–up–down spin structure along the b-axis.

As shown in figure 1, each of the spectra between 13.7 and 13.8 T has a double peak structure around the upper peak of the resonance line of the B site. Furthermore, the shifts of the two peaks almost coincide with the upper peaks for commensurate (13.603 T) and incommensurate (13.905 T) phases. Actually, this double peak structure is reproduced well

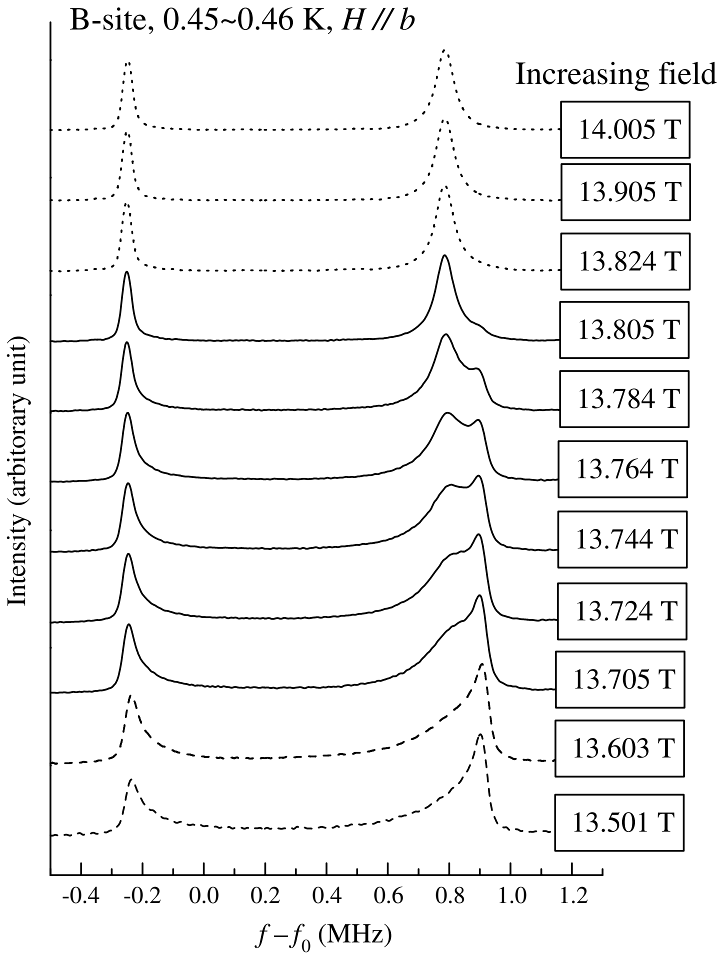

Figure 1. ^{133}Cs NMR spectra of the B site obtained at 0.45–0.46 K under a field applied along *b*-axis. The abscissa shows the shift in frequency. The applied field is increased from 13.5 to 14.0 T. The spectra in the incommensurate and commensurate plateau phases are drawn by dashed and dotted lines. Each spectrum with a solid line shows a coexistence of the two phases.

by mixing the spectra of commensurate (13.603 T) and incommensurate (13.905 T) phases in a suitable percentage. The percentage of commensurate and incommensurate phases is obtained as shown in figure 2 from the best fit to the experimental results. A gradual development of the incommensurate–commensurate transition is clearly observed as a function of the field. Thus, coexistence of the two phases is strongly suggested.

We also found a hysteresis of the spectrum for this field-induced transition. Figure 3 shows that the spectrum of 13.724 T in increasing field is clearly different from that at the same field for decreasing field. Similar hysteresis and coexistence are also found at the higher end of the plateau, which will be discussed elsewhere.

We found hysteretic behaviour at each end of the plateau in the field dependence of the nuclear spin–lattice relaxation rate T_1^{-1} of ^{133}Cs shown in figure 4. Here, T_1^{-1} is measured at 0.5 K for the lower frequency peak of the resonance line of the B site which has an almost constant shift as shown in figure 1. Our results for the spectrum and T_1^{-1} provide

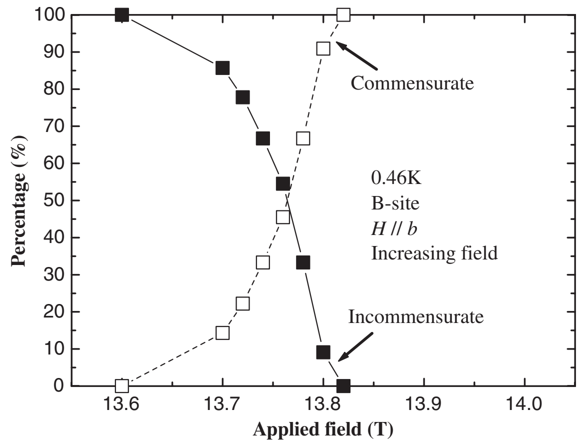

Figure 2. Field dependence of the percentage of the commensurate and incommensurate phases estimated from the fitting to the spectrum between 13.7 and 13.8 T. The incommensurate phase at lower field is gradually switched to the commensurate plateau phase.

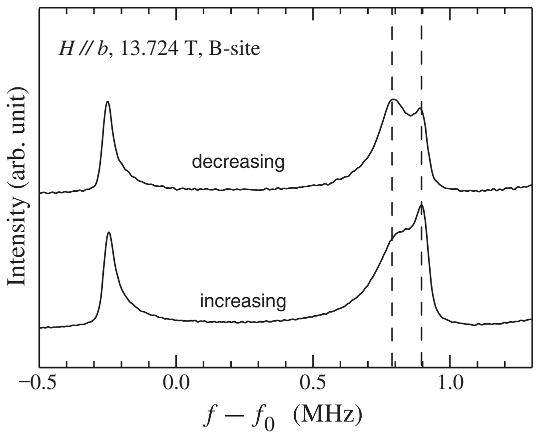

Figure 3. ^{133}Cs NMR spectra of the B site obtained at 0.46 K under the field of 13.724 T obtained while increasing and decreasing the field applied along the *b*-axis. The abscissa shows the shift in frequency. The dashed lines are drawn to guide the eyes.

evidence of first order phase transitions at both ends of the plateau region. Thus, T_1^{-1} abruptly changes around the phase transition as the field is increased. It is found that the change of the value of T_1^{-1} in the hysteresis around the upper critical field (15.3–15.4 T) was rather small as compared with that around the lower critical field (13.6–13.82 T). This probably means that these two transitions are different to each other. Such a difference was also suggested from the magnetocaloric effect measurement. [9] Notice that T_1^{-1} gradually increases with increasing field above 14.5 T. In the plateau field region, T_1^{-1} decreases steeply with decreasing temperature [8], which presumably suggests the presence of an energy gap. Then, our results suggest that low energy magnetic excitations are modified by the magnetic field while a commensurate spin structure is retained. It should be noted here that the incommensurate phases below and above the plateau phase resemble each other from the viewpoint of the NMR spectrum and T_1^{-1}.

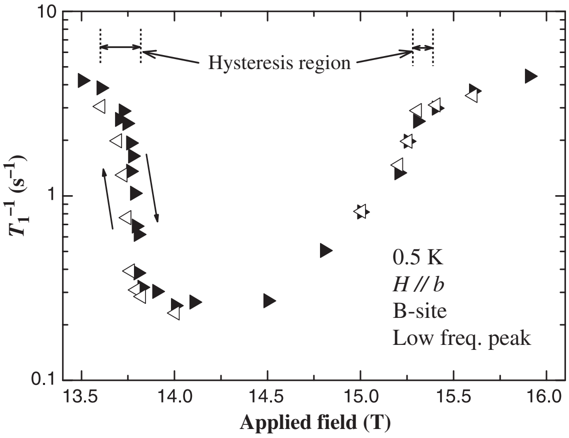

Figure 4. Field dependence of T_1^{-1} for ^{133}Cs at 0.5 K for the resonance line at lower frequency of the B site. The solid triangles and open triangles indicate the data for increasing and decreasing field, respectively. The dotted lines indicate ranges in which hysteresis of T_1^{-1} is observed.

In summary, we have found clear evidence of the first order phase transition between incommensurate and commensurate (plateau) phases in the ^{133}Cs NMR spectrum and its relaxation rate T_1^{-1} for the distorted triangular antiferromagnet Cs_2CuBr_4. We also found an asymmetric behaviour within the plateau region in T_1^{-1}. Detailed experimental results and a discussion will appear elsewhere.

Acknowledgments

This work was partly supported by a Grant-in-Aid for Young Scientists (B), No. 17740221, from the Ministry of Education, Culture, Sports, Science and Technology of Japan.

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