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2005 J. Phys.: Condens. Matter 17 1335

(http://iopscience.iop.org/0953-8984/17/8/012)

INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **17** (2005) 1335–1340 doi:10.1088/0953-8984/17/8/012

The magnetic spiral in the frustrated antiferromagnet $RbCuCl_3$ studied by means of neutron diffraction

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Received 13 December 2004, in final form 20 January 2005
Published 11 February 2005

Abstract

A neutron diffraction study was performed to study the spiral spin arrangement of $RbCuCl_3$ as a function of temperature and magnetic field. We clearly observe that the spiral lengthens with increasing temperature up to $T_N = 19.3$ K. A shortening of the spiral occurs when a magnetic field is applied within the plane of spin rotation. No change in the propagation vector is observed with the field applied perpendicular to the spin rotation plane. In addition we measured the temperature dependence of the magnetization and close to T_N we determine critical exponents $\beta = 0.238(4)$ and 0.26(1) at zero field and at 13.5 T, respectively.

1. Introduction

$RbCuCl_3$ is closely related to the ABX_3 compounds which crystallize in the hexagonal space group $P6_3/mmc$. The magnetic B^{2+} transition metal ions in these systems are located on a triangular sublattice in the *ab*-plane with a weak antiferromagnetic coupling J_{ab} between nearest neighbours. A much stronger coupling J_c is present between nearest neighbours along the *c*-axis where the B^{2+} ions form linear chains. Therefore, most ABX_3 compounds represent quasi-one-dimensional magnets. Many of these systems have been investigated intensively in the past [1]. Particular interest was focused recently on those systems where frustrated antiferromagnetic interactions in the *ab*-plane give rise to unusual magnetic properties.

One of these magnetically frustrated systems is $RbCuCl_3$ which shows two structural phase transitions. At 339 K it changes from the hexagonal to an orthorhombic structure ($Pbcn$). Below 260 K it becomes monoclinic ($C2$). Recent neutron diffraction work indicated an antiferromagnetic order below 19 K with a propagation vector $Q = (0.2993, 0.2993, 0)$ corresponding to a structure with a spin turning angle of about 108° (figure 1), [2]. The indexing used here refers to the high temperature hexagonal crystal structure setting. The angle of 108° is below the 120° for the regular triangular antiferromagnet. One can attribute this difference to the orthorhombic distortion of the lattice since then only two of the three

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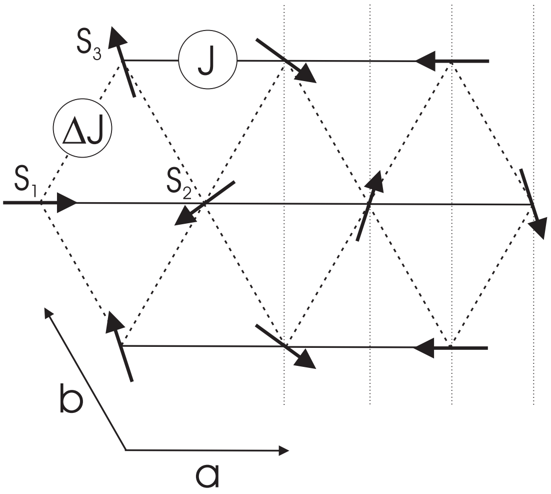

Figure 1. Incommensurate spin structure in the basal plane. Spins S_1, S_2, and S_3 define the distorted triangular spin arrangement. J and $\Delta \cdot J$ are exchange interactions. Also given are lattice constants a, b using an approximate description of the orthorhombic structure by means of a hexagonal setting.

couplings on the deformed triangular plaquette remain equivalent. Recently, high field specific heat measurements revealed two anomalies in a small temperature window near 19 K [3]. This result was attributed to the existence of an intermediate phase between the paramagnetic and the frustrated antiferromagnetic phase. Our work presented here aims to study in detail the temperature and field behaviour of the antiferromagnetic structure and to look at this intermediate phase by means of neutron diffraction.

2. Experimental details

The diffraction experiments were carried out at BENSC using the E1 triple-axis spectrometer with removed analyser. The wavelength was 2.4 Å provided by a pyrolytic graphite monochromator. Second-order contamination $\lambda/2$ was suppressed by a pyrolytic graphite filter and the collimations α_1, α_2, α_3, and α_4 were set to open, 20′, 40′, and open, respectively. The samples were single crystals of $RbCuCl_3$ prepared from a melt [2]. The linear dimensions of the measured crystals were about 5–8 mm. The field dependence of the magnetic structure was measured up to 14.5 T using the vertical superconducting magnet VM1. Temperatures were varied between 2 and 20 K. The crystal was mounted in the magnet in two different orientations. Either the scattering plane was chosen as $(h\ h\ l)$ so that the field is inside the ab-plane or the scattering plane was $(h\ h\ 0)$ now with the field pointing along the c-axis. Nuclear reflections of the forms of planes $\{2\ 2\ 0\}$ and $\{0\ 0\ 2\}$ were used to carry out the appropriate orientations and scans were always performed along $(h,\ h,\ 0)$ in reciprocal space to measure the magnetic diffraction.

3. Measurements and data analysis

At zero field we recorded the temperature dependence of the magnetic satellite peak. Figure 2 shows the temperature variation of the peak position. We see a shift in the position of the

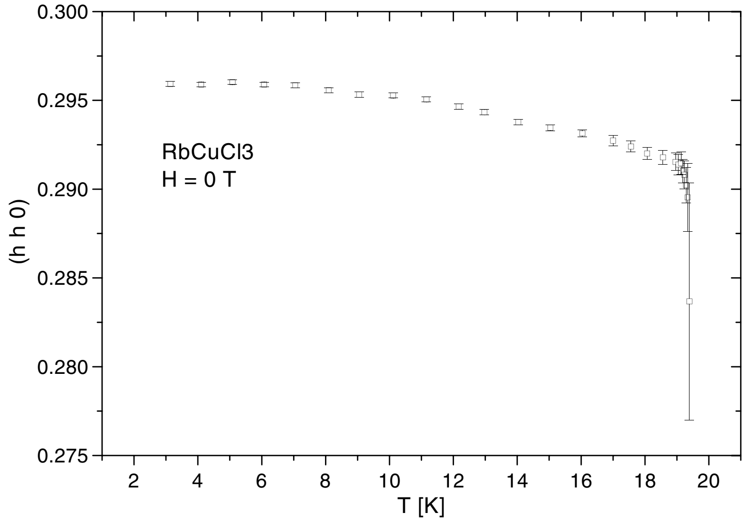

Figure 2. Temperature dependence of the magnetic propagation vector $Q = (h,\ h,\ 0)$.

(0.297 0.297 0) reflection at 2 K corresponding to a reduction of the propagation vector with increasing temperature. The peak intensity vanishes at about 19.3 K. Extinction effects are assumed to be small and are not considered since the intensities of the magnetic reflections are weak. Our experiments indicate a continuous phase transition. We have calculated the reduced magnetization $m = M/M_0$ from the measured integrated intensity I as $m(T) = (I/I(2K))^{1/2}$ and we have fitted m to a power law $m = b * ((T_N - T)/T_N)^{\beta}$ in a reduced temperature range $t \leqslant 2 \times 10^{-1}$ (figure 3). We obtain a critical exponent $\beta = 0.238(4)$, a critical amplitude $b = 1.17(1)$, and the transition temperature is determined as $T_N = 19.35(1)$ K. No significant critical scattering is present around T_N. A similar temperature dependent measurement was done at an in-plane field of 13.5 T. Again we find a shift in the position of the satellite peak and the fit of m to the critical power law yields $\beta = 0.26(1)$ and $T_N = 18.93(2)$ K.

Field dependent measurements of the magnetic reflection between 0 and 12 T at 2 and at 19 K show a small shift in the peak position related to an increase of the propagation vector with growing field strength. Besides applying the field inside the *ab*-plane we have also studied the case with the field along the *c*-axis. No field dependence of the peak position is observed now. In all cases we have carefully checked possible variations of nuclear reflections on field and temperature. Below 20 K and up to 14.5 T no field or temperature variations could be observed and it was sufficient to use the orientation matrix determined at 2 K and at zero field.

4. Discussion

The magnetic spiral observed earlier by means of neutron diffraction can be attributed to the orthorhombic distortion of the hexagonal structure (figure 1). The Hamiltonian of the in-plane magnetic interactions in $RbCuCl_3$ on the triangular plaquette is written as

$$H_{ab} = \Delta \cdot J(\vec{S}_1\vec{S}_3 + \vec{S}_2\vec{S}_3) + J\vec{S}_1\vec{S}_2 = J/2 \cdot (\vec{S}_1 + \vec{S}_2 + \Delta\vec{S}_3)^2 - \tfrac{1}{2}J\vec{S}^2(2 + \Delta^2) \qquad (1)$$

where the S_i denote the spins, J is the antiferromagnetic coupling, and Δ takes into account the orthorhombic distortion yielding a spatially anisotropic intraplane exchange so that only two of the couplings remain equivalent. The complete Hamiltonian including the ferromagnetic

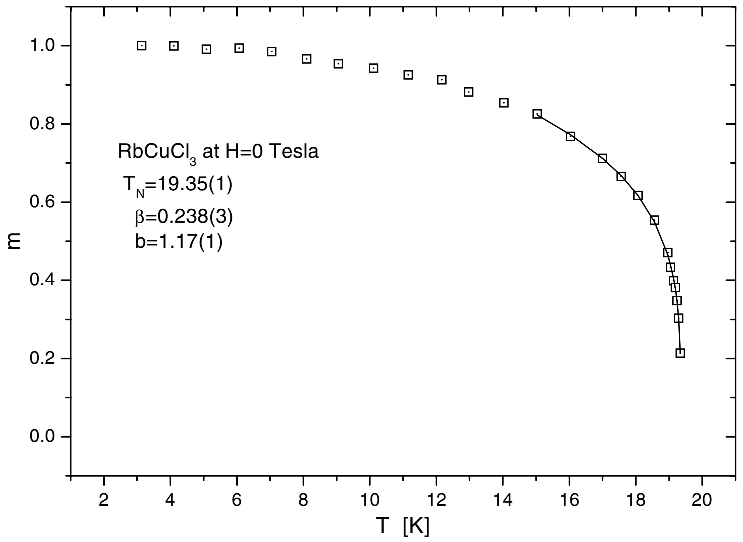

Figure 3. Temperature dependence of the magnetization. The full curve indicates the fit of $m(T)$ to a critical power law behaviour (see the text).

intrachain interaction and the easy plane anisotropy can be found elsewhere [4]. Minimizing (1) by putting the first term of the second expression equal to zero yields

$$\cos(2\pi Q) = -\Delta \tag{2}$$

which relates the angle $\varphi = 2\pi Q$ between adjacent spins to the parameter Δ.

First we will discuss the observed field behaviour of the magnetic propagation vector Q. With the longitudinal field parallel to c we find no change in the propagation vector and we only notice a tiny decrease in the magnetic satellite intensities. This behaviour is expected for a system with the spins lying in the ab-plane. At 14 T our field is limited to about $0.2H_s$ as the saturation field H_s is near 66 T [4]. Our observations are consistent with the presence of an umbrella-type structure in the field where all spins are lifted out of the ab-plane towards c and the propagation vector remains unchanged. Magnetic diffraction intensity is then transferred from the satellite peaks to the reciprocal lattice positions of the nuclear reflections due to the formation of a ferromagnetic component in the structure. Considering the linear increase of the magnetization with field [4] we estimate a lifting of spins towards c by 12° at the highest field strength available. This corresponds only to a decrease of the magnetic satellite intensity by 4% in agreement with our measurement.

More interesting is the case of the transverse field. Here magnetization measurements show a jump in $M(H)$ around 21 T at 1.3 K indicative of a phase transition [4]. From neutron diffraction in fields up to $H = 12.5$ T $\cong 0.2H_s$ we see a clear increase of the propagation vector (figure 4). A comprehensive theoretical study of the magnetic structures of $RbCuCl_3$ in a transverse field was reported recently [5]. These examinations are based on a model Hamiltonian in a classical approximation. Mainly three different regions of field strengths have to be considered. At small fields an incommensurate structure with wavevector in the layer should be optimal. In this low field region with $H < 0.4H_s$ the field induced modulations of the regular spiral were calculated numerically. Figure 4 shows the field variation of the Q-vector obtained in [5] compared with our experimental findings. There is qualitative agreement with respect to the increase of the Q-vector with growing field. Up to 6 T $\cong 0.1H_s$ the Q-

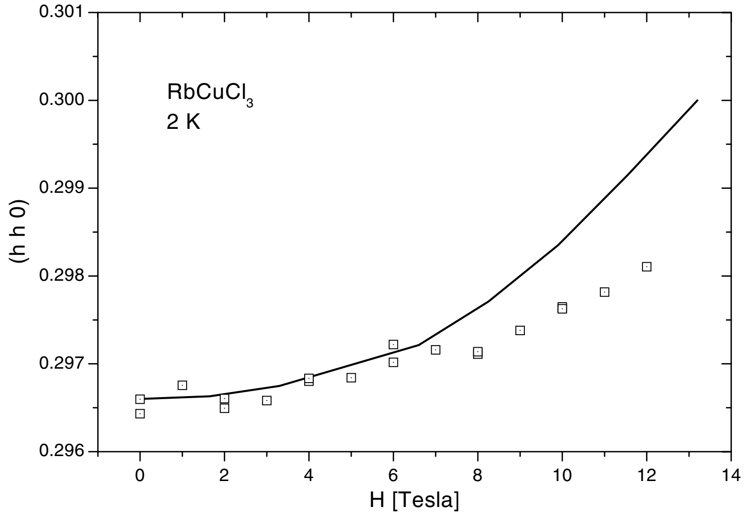

Figure 4. Field variation of the magnetic propagation vector $Q = (h, h, 0)$. The full curve indicates calculations of $Q(H)$ [5] which has been shifted to agree at zero field with the values obtained experimentally.

dependence agrees quantitatively while the Q-vector is well below the theoretical prediction at higher fields. The different magnetic structures calculated for intermediate fields as well as the incommensurate fan structure predicted for the high field region cannot be tested since we are limited by the available field strengths. It will be noted that the calculated field dependence of the magnetization reproduces well the jump in $M(H)$ reported in [4].

Now we will discuss the observed temperature behaviour of the magnetic satellite peaks. Figure 2 shows that $\mathrm{d}Q/\mathrm{d}T$ is nearly zero at the low temperature side; then Q decreases continuously and falls off more rapidly quite close to T_N. The analysed decay in the satellite intensity indicates a continuous phase transition (figure 3). The critical exponents β of about $1/4$ determined for the temperature behaviour of the order parameter are very close to values observed in many other ABX$_3$ systems as well. The comparison of our field dependent data to the specific heat measurements shows that our transition corresponds to the large anomaly in the specific heat [3]. The difference of about 0.4 K in the absolute value of the transition temperature between neutron and specific heat data has to be attributed to calibration errors. The neutron data indicate a continuous magnetic transition and we cannot find evidence for an intermediate phase. The absence of significant critical scattering is similar to what has been observed in CsCuCl$_3$. There only very weak intensity from critical scattering could be detected. The critical exponent $\beta = 0.25$ found earlier for CsCuCl$_3$ [6] agrees well with the one found for RbCuCl$_3$.

No indication was found for an intermediate magnetic phase observed in the specific heat. The temperature region for this intermediate phase amounts only to a few tenths of a degree and it increases with applied field. We propose that chiral effects can cause it. Let us refer to theoretical studies of the fully frustrated two-dimensional spin systems on a triangular lattice. The antiferromagnetic coupling on the triangle results in the 120° structure and the order parameter for the ground state corresponds to the mean magnetization of each of the three sublattices. Since a chirality with pseudospin + or − can be attributed to each triangle one can consider a second-order parameter of Ising type as well. Theoretical analyses now predict [7, 8] that the two transitions occur at different temperatures. This should show up in two anomalies for the specific heat as were observed experimentally [3]. In our neutron diffraction experiment we probe solely the '120°' (108°) structure and observe magnetic Bragg intensities at the rlp around (0.3 0.3 0), i.e. we measure the mean sublattice magnetization. Usually chiral effects are not observable with unpolarized neutrons. Only under special circumstances like those in $CsCuCl_3$ they can become accessible [9]. There we observed a commensurate incommensurate phase transition of the spin arrangement along c with rising field. This was accompanied by a change in the population of macroscopic domains with different chirality on the triangular lattice in the ab-plane. For $RbCuCl_3$ the two different types of chirality are not distinguishable and we cannot get any information on the chiral order either on a macroscopic or on a microscopic scale. This means that the onset of chiral order is not seen by means of unpolarized neutron diffraction; however, it can be observed in the specific heat.

In conclusion our experiments show that the propagation vector of the incommensurate magnetic structure in $RbCuCl_3$ changes with field as well as with temperature. The field variation is in qualitative agreement with theoretical predictions [5]. The weaker dependence of q on the field found experimentally is similar to earlier observations at $CsCuCl_3$. The phase transition at about 19.3 K is second order with a critical exponent $\beta = 1/4$ which is found in other frustrated ABX_3 systems as well [1]. The temperature and field dependent transition to the paramagnetic state corresponds to the sharp anomaly observed in specific heat measurements [3]. An intermediate magnetic phase indicated by a weak shoulder in the specific heat has not been observed in our neutron diffraction work. In the case where this transition is related to the order in chirality it cannot be observed by means of magnetic diffraction with unpolarized neutrons.

Acknowledgments

We thank U Schotte and K D Schotte for fruitful discussions and H Tanaka for providing us with the single crystals.

References

[1] Collins M F and Petrenko O A 1997 *Can. J. Phys.* **75** 605 and references therein

[2] Reehuis M, Feyerherm R, Schotte U, Meschke M and Tanaka H 2001 *J. Phys. Chem. Solids* **62** 1139

[3] Pérez-Willard F, Faißt A, Wosnitza J, v Löhneysen H, Schotte U and Tanaka H 2000 *Eur. Phys. J.* B **15** 247

[4] Marayuma S, Tanaka H, Narumi Y, Kindo K, Nojiri H, Motokawa M and Nagata K 2001 *J. Phys. Soc. Japan* **70** 859

[5] Jacobs A E and Nikuni T 2002 *Phys. Rev.* B **65** 174405

[6] Schotte U, Stüßer N, Schotte K D, Weinfurter H, Mayer H M and Winkelmann M 1994 *J. Phys.: Condens. Matter* **6** 10105

[7] Miyashita S and Shiba H 1984 *J. Phys. Soc. Japan* **53** 1145

[8] Noh J D, Rieger H, Enderle M and Knorr K 2002 *Phys. Rev.* E **66** 026111

[9] Stüßer N, Schotte U, Hoser A, Meschke M, Meissner M and Wosnitza J 2002 *J. Phys.: Condens. Matter* **14** 5161