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Fluctuation-induced phase in $CsCuCl_3$ in a transverse magnetic field: experiment

U Schotte†, A Kelnberger and N Stüsser
Hahn-Meitner-Institut, BENSC (Neutron Scattering Group), D-14109 Berlin, Germany

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Abstract. The new magnetic phase of $CsCuCl_3$ recently found by specific heat measurements has been investigated by means of neutron diffraction. This new phase is found very close to the critical temperature and it widens to a few tenths of a degree Kelvin in very high magnetic fields. The availability of a new strong steady-state magnet with a nominal strength of 14.5 T provided us with the opportunity to investigate the magnetic structure. We found that the new phase is also incommensurate, but that it no longer has the 120° triangular antiferromagnetic structure. A proposal for the structure derived from a Landau theory put forward in the following theory paper by Jacobs and Nikuni is compared to the experimental results. The order of the transitions observed will be discussed, as well as our failure to observe a predicted commensurate phase.

1. Introduction

We present details of the newly discovered magnetic phase of the hexagonal perovskite $CsCuCl_3$ in an external field which is orthogonal to the crystal *c*-axis. This new phase was identified first in specific heat measurements and then in the magnetization investigations (Werner *et al* 1997) close to the Néel temperature T_N.

$CsCuCl_3$ is interesting because it belongs to the group of 'frustrated' magnetic systems with a triangular lattice of antiferromagnetically coupled $S = 1/2$ spins of Cu^{2+} in the *a*–*b* plane ('antiferromagnetic 120° structure'); the dominating magnetic interaction along the *c*-axis is ferromagnetic and there is an additional Dzyaloshinsky–Moriya (DM) interaction. From both of these there result incommensurate magnetic helices winding around the *c*-axis with a small turn angle such that the repeat length is about 70 layers. The DM interaction forces the spins to lie almost flat in the *a*–*b* plane, so this spin system is approximately an XY-system. The ratio of inter-plane to intra-plane exchange is about 6; therefore the system cannot be called a quasi-one-dimensional one. Since the mean-field value of T_N is about 35 K while the actual value is near 10.7 K, strong deviations from mean-field behaviour are expected due to frustration and fluctuation effects of the small spin.

Large single crystals can be produced from solution. Therefore many detailed experimental results are currently available. We only mention the most striking results. In external fields parallel to the *c*-axis ($H > 5.4$ T near T_N), the system undergoes a flop-like first-order phase transition from the so-called umbrella structure to a 'collinear' structure with two sublattice spins in the same direction and the spins and field in a common plane (Nikuni and Shiba 1993, Schotte *et al* 1994, Motokawa and Arai 1995). Not only is this

† Also at: Institut für Mineralogie, Universität Kiel, Germany; e-mail: schotte@hmi.de.

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fluctuation-triggered transition of interest, but so also is the validity of the chiral universality class, or, rather, the controversy surrounding it (Lee *et al* 1984, Kawamura 1985, 1992, Zumbach 1993, Antonenko and Sokolov 1994) and the checking of the predicted critical exponents. The specific heat measurements indicate that at zero field the phase transition is either tricritical or weakly first order (Weber *et al* 1996). This is also in agreement with recent computer simulation results obtained using a model which resembles $CsCuCl_3$ (Plumer and Mailhot 1997, Loison and Schotte 1998). In non-zero fields, the large values of α found are closer to those predicted by Kawamura, with not much difference between the two phases. From the temperature dependence of the magnetic diffraction peaks, β stays near 0.25, which holds for the tricritical transition as well as for Kawamura's prediction.

Specific heat measurements in fields making different angles with respect to the c-axis show qualitative changes in the phase diagrams near T_N: while for $\boldsymbol{H} \parallel \boldsymbol{c}$, $T_N(H)$ falls with temperature, it rises for $\boldsymbol{H} \perp \boldsymbol{c}$. Common to all cases is a first-order phase transition to a new spin structure which apparently reaches paramagnetic saturation via a second-order transition (Wosnitza *et al* 1998).

The basic Hamiltonian, already described above, is stated as follows (Tanaka *et al* 1992):

$$H = -2J_0 \sum_{in} (\boldsymbol{S}_{\mathrm{i}n} \cdot \boldsymbol{S}_{in+1} + \eta(S^x_{\mathrm{i}n} S^x_{in+1} + S^y_{in} S^y_{in+1})) + J_1 \sum_{i \neq jn} \boldsymbol{S}_{in} \cdot \boldsymbol{S}_{jn}$$
$$- \sum_{in} \boldsymbol{D} \cdot (\boldsymbol{S}_{in} \times \boldsymbol{S}_{in+1}) - g\mu_B \sum_{in} \boldsymbol{H} \cdot \boldsymbol{S}_{\mathrm{i}n} \tag{1}$$

where n numbers the spins along the chains, i and j number the chains, and $J_0 = 28$ K, $J_1 = 4.9$ K and $D/J_0 = 0.18$.

The DM interaction effectively tends to spread the spins apart, working against the ferromagnetic exchange and thus producing the incommensurate spiral.

For crystal symmetry reasons, the vector $\boldsymbol{D}$ of the DM interaction must be, at least on average, parallel to the c-axis; in fact $\boldsymbol{D}$ follows the structural spiral of the Jahn–Teller-distorted chloride octahedra.

From the structure determination, the spins are known to be slightly canted out of the *a*–*b* plane (Adachi *et al* 1980). The very small exchange anisotropy η, as verified by ESR experiments, and the DM interaction each effectively produce an easy-plane anisotropy. These anisotropies are relatively small, which has led Nikuni and Shiba (1993) to consider fluctuations as a source for the triggering of the spin-flop transition in an external field $\boldsymbol{H} \parallel \boldsymbol{c}$.

Also, for our investigations with strong fields in the *a*–*b* plane, near the critical temperature the canting and the anisotropies are considered negligible, since, in that region, fluctuations are known to dominate.

Without these anisotropies, the ground-state energy per spin, up to a constant, can be written as

$$E_0/N = J_1\left(\boldsymbol{S}_1 + \boldsymbol{S}_2 + \boldsymbol{S}_3 - \frac{g\mu_B \boldsymbol{H}}{6J_1}\right)^2. \tag{2}$$

This is taken as approximately valid also for $\boldsymbol{H} \perp \boldsymbol{c}$.

While it is plausible that in an external field parallel to the c-axis the spirals could keep their winding sense and repeat length while the sublattice spins change their relative angles, this cannot be so when the field is in the *a*–*b* plane: naively one would expect the spirals to stretch out until all of the spins were parallel to the field with the result that the magnitude of the spiral wave vector $\boldsymbol{Q}$ would go to zero as the field and/or the temperature rose. Mean-field calculations do indeed predict a commensurate phase between the incommensurate phase and paramagnetic saturation at high fields (Jacobs *et al* 1993). There is now experimental evidence that this happens in very high fields and for low temperatures: at 4.5 K, $\boldsymbol{Q}$ continuously goes to zero when the field reaches 17 T (Nojiri *et al* 1998). However, before this happens, $\boldsymbol{Q}$ seems to develop a plateau (Motokawa and Arai 1995). This overall steplike behaviour of $\boldsymbol{Q}(\boldsymbol{H})$ has been explained as due to fluctuations by Nikuni and Jacobs (1998).

Close to T_N, the situation becomes even more complicated as a result of the appearance of the new phase and the observation that T_N rises with rising field. According to our neutron results, to be discussed in detail below, the new phase is incommensurate, with a Q-value again larger but not too far away from the zero-field value.

2. The experimental set-up and results

The neutron diffraction experiments were carried out on the triple-axis spectrometer E1 at the Berlin Neutron Scattering Centre. The wavelength $\lambda = 2.42$ Å was selected by Bragg reflection at a pyrolytic graphite monochromator for the incoming beam and the analyser was adjusted to measure the elastic part of the scattering. The samples were mounted in the new high-field superconducting magnet VM1 built by Oxford Instruments, which with a maximum field of 14.5 T is actually the strongest steady-state magnet available for neutron scattering. The main characteristics are a split coil with a bore of 20 mm and a diameter of 15 mm. The vertical opening angle for collecting scattered neutrons amounts to $\pm 2^\circ$. The available temperature range is from 300 K to 1.5 K and this can be extended down to 100 mK by using a dilution insert. The experiments described below were performed in fields not higher than 13 T.

$CsCuCl_3$ samples about 1 cm^3 in size—the same ones as were studied earlier (Schotte *et al* 1994)—were mounted with the $(h\,h\,l)$ zone in the horizontal scattering plane and the field was along the $h\,0\,0$ direction.

First the temperature and field dependences of the $(\frac{1}{3}\,\frac{1}{3}\,Q)$ reflection were investigated, by measuring this peak at fixed T and for different values of H, with the aim of finding the transition to a commensurate phase as predicted by theory and found previously for a temperature of 4.5 K.

The results are shown in figure 1: at zero field the value of Q is 0.085. For T fixed at 10.34 K, Q falls in a rising field to about 0.048 at 11.75 T. After the next H-step, at 12 T the peak has disappeared; the transition appears quite abrupt, in that the peaks do not broaden or lose intensity on approaching 12 T. At $T = 9.65$ K, Q drops to 0.047 at 13 T; with no field higher than 13 T applied, there is no experimental indication of how far away this is from the paramagnetic phase: again all of the magnetic peaks were narrow (within the limits of the instrumental resolution) and characteristic for the incommensurate phase of distorted spirals with the equivalent sublattice spin phase shifted along the c-axis by $2\pi/3$; this structure will be called IC1.

Also shown in figure 1 are tentatively inserted theoretical curves ($\alpha_1 \sim T$ is a Landau parameter; for details, see the theory paper by Jacobs and Nikuni (1998), which is the following article); these were included to show that the data fit to the theoretical expectation for the behaviour of Q in a field which changes with T—namely, the higher the value of T, the sharper the drop. In order to scale the theoretical curves to fit into figure 1, $H_s = 30$ T was used; however, we did not intend to imply any quantitative agreement.

Both in zero field and at 10.5 T (figure 2), the peak intensities were measured as a function of temperature in an effort to extract the critical exponent β. For both cases, β

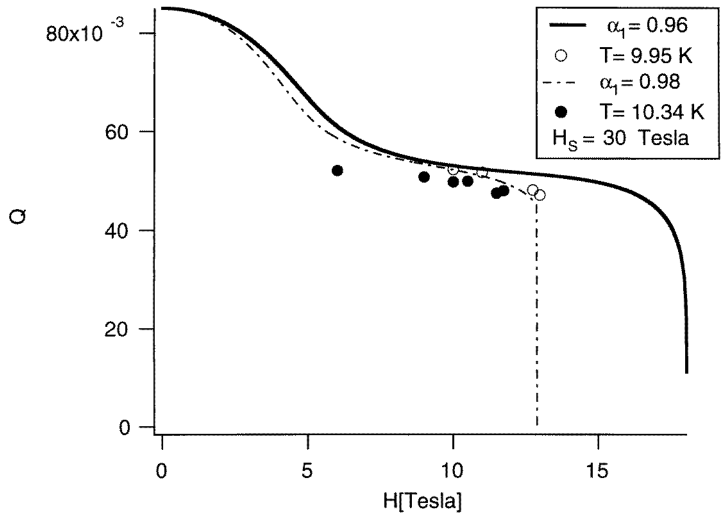

Figure 1. Experimental results for the spiral pitch in high fields close to the transition to paramagnetism. For $T = 10.34$ K, the sharp drop beyond 11.75 T indicates the sudden disappearance of the magnetic peak. For $T = 9.65$ K, the paramagnetic state could not be reached. The same behaviour is mirrored by the theoretical curves scaled into this figure; for details and the parameters $\alpha_1 \sim T$, see the following theory paper by Jacobs and Nikuni.

stays near 0.25; it is somewhat smaller in the high field, similarly to the $\boldsymbol{H} \parallel \boldsymbol{c}$ situation. On the one hand, it is interesting to see how stable this value is, but, on the other hand, this cannot mean much, since, in the light of the results below, the data points close to T_N lie in or skip over the new incommensurate phase, so one cannot say anything about the

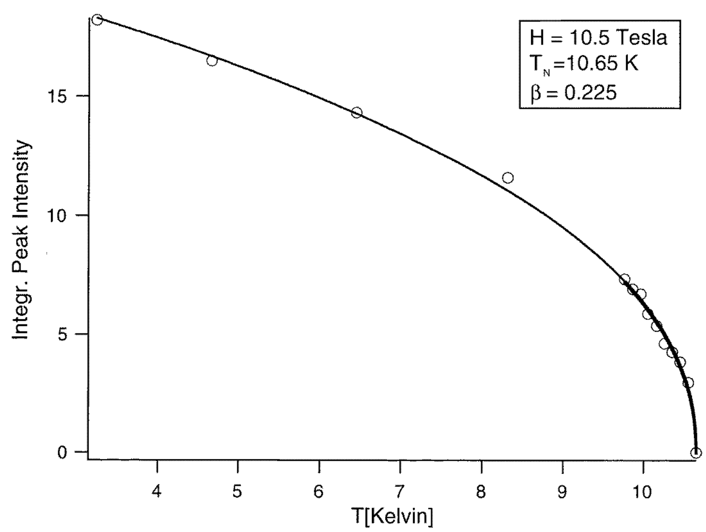

Figure 2. The integrated peak intensity of the magnetic peak at $(\frac{1}{3}\ \frac{1}{3}\ 0.055)$ in a field of 10.5 T fitted with a $(T - T_N)^{2\beta}$-law. The value 0.225 comes from a fitting close to the critical temperature, marked by the thicker line.

region very close to T_N where a critical exponent would be meaningful.

The major part of our investigation was that concerned with the l-scans near $(\frac{1}{3}\,\frac{1}{3}\,l)$ with $-0.15 \leqslant l \leqslant 0.15$ in the region where the new phase was to be expected: H and T for these scans are marked with several kinds of 'round symbol' in the phase diagram in figure 3. Also, results from other experiments, together with a 'guide to the eye' to the border of the paramagnetic region, are indicated.

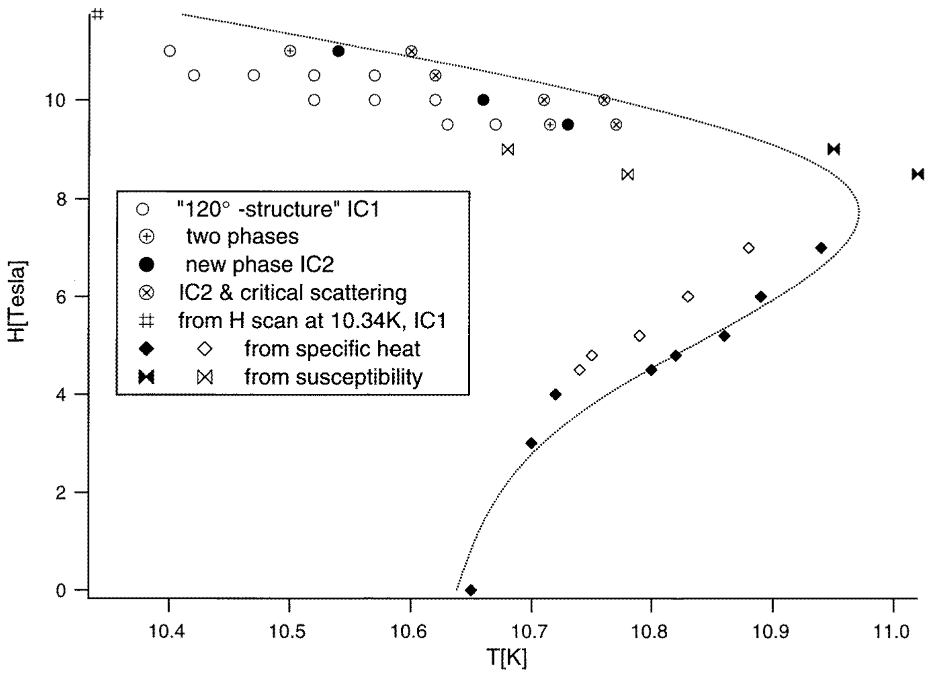

Figure 3. Part of the phase diagram where the new phase IC2 could be localized by means of specific heat, susceptibility and neutron diffraction measurements; only the data indicated by the 'round symbols' will be discussed below. Note that the new phase is only a few tenths of a degree Kelvin wide.

In figures 4–7 typical results are presented; the scans at 9.5 T were chosen—others of comparable type would be indistinguishable from these, because in the region investigated the variations of Q, the linewidths and the peak ratios stay within the experimental errors.

The offset of $l = 0$ from the centre between the peaks is due to the non-perfect orientation of the crystal sample and has no influence on the interpretation. In addition to the data points, the residuals that are the differences between the data and the peak fits are shown.

There are four distinct regimes distinguishable in these scans, with rising temperature.

(1) Close to the borderline of the new phase (see figure 4), four peaks characterize the IC1 structure: the main magnetic peaks are at $\pm Q$ and the first harmonics at $\pm 2Q$; here and for all of the scans in the region described, $Q \approx 0.052$; this indicates a repeat length of about 115 spins along $\boldsymbol{c}$.

(2) In the 'two-phase regime' (see figure 5), seven peaks can be fitted: the IC1 structure, still present, plus three new peaks, with a central component and side peaks near $\pm Q_{\mathrm{new}} \approx \pm 0.074$; this phase will be called IC2. The higher harmonics of the latter would

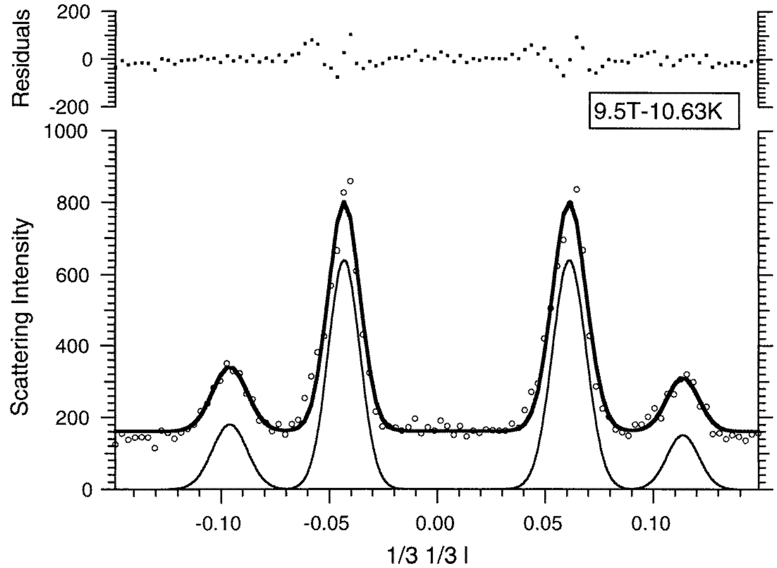

Figure 4. The normal magnetically ordered state of $CsCuCl_3$ in an external field: the smaller peaks are higher harmonics due to the distortion of the spirals and the spiral pitch Q has dropped from 0.085 (at $H = 0$) to 0.054—that is, the spirals have started to stretch out.

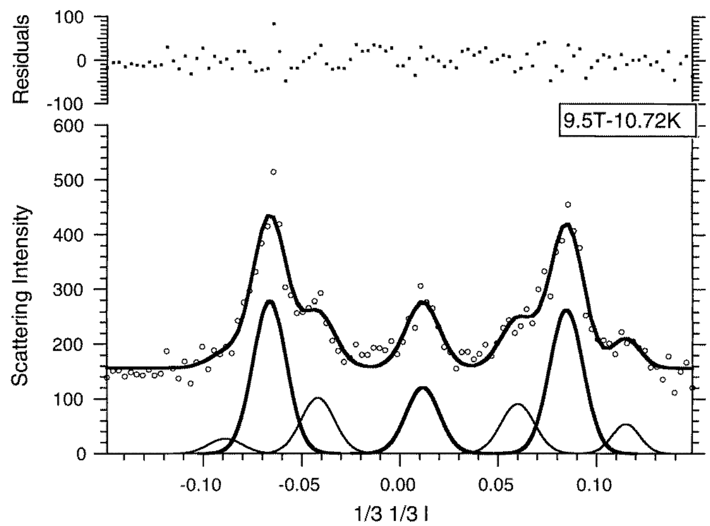

Figure 5. Peaks for the two phases IC1 and IC2 coexisting, the central and side peaks at $Q_{\mathrm{new}} = 0.074$ of the new phase being already somewhat stronger than those of the 120° structure.

be just at the border of the scan range and probably too low in intensity to be detected. The coexistence of the two phases points to a first-order phase transition, for which hysteresis effects are to be expected; however, they have not been looked for on this occasion.

(3) The peak structure of the new phase is shown in figure 6. It is improbable that these peaks indicate a commensurate structure with the side peaks as first harmonics, the side

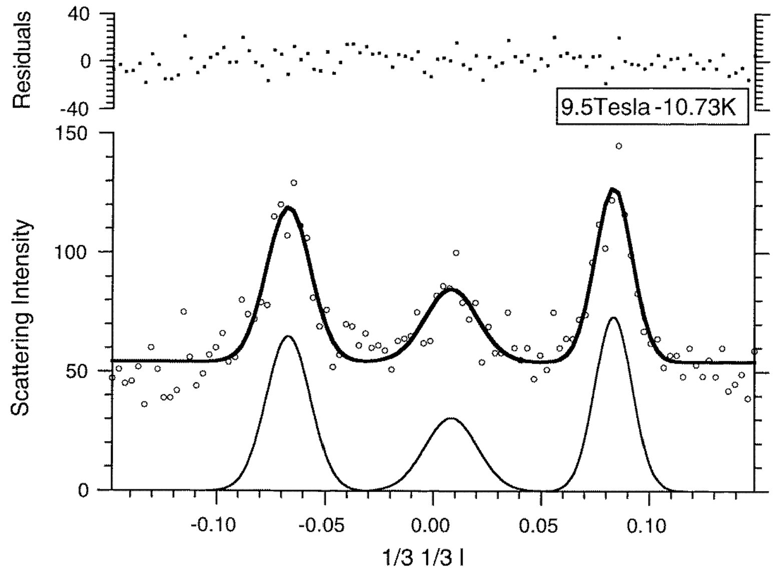

Figure 6. Peaks for the new phase. Note that the scattering intensity has reduced, the monitor/detector settings being the same for all of the experiments described, and that the peaks are broader than those of figures 4 and 5.

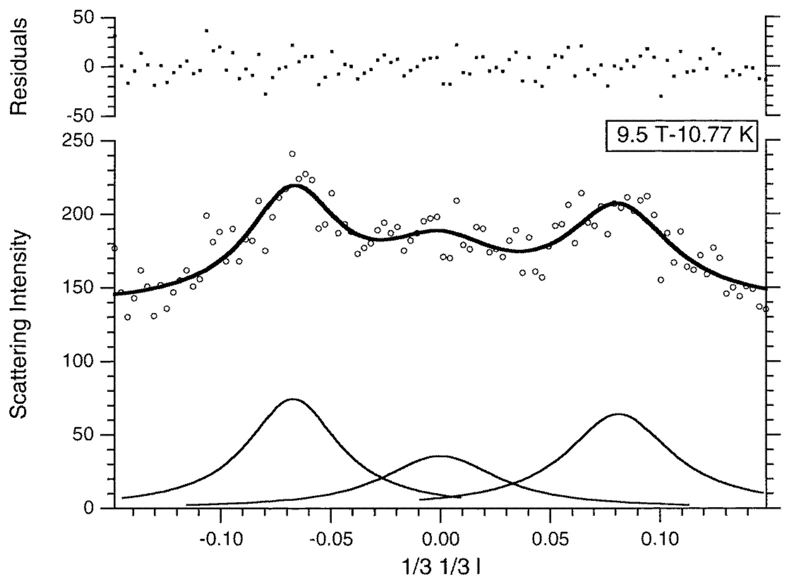

Figure 7. Peaks for the new phase with critical scattering: $Q_{\mathrm{new}} = 0.074$ does not change measurably, but the peaks just 'dissolve' in the background. Here the Voigt fit gives a Lorentzian halfwidth of 0.0294.

peaks being twice as high as the central one. From experience with the $\boldsymbol{H} \parallel \boldsymbol{c}$ case, for which the peak structure looked quite similar, we know that at this point one has to discard the model of a 120° structure with three equivalent sublattices. A model of the IC2 phase will be described below.

(4) As the critical temperature is approached, the peaks of the new structure do not shift towards zero, but the peaks broaden and disappear into the background (see figure 7); apparently critical scattering is observed, which points to a second-order phase transition. The width, taken as dominantly Lorentzian according to the Voigt fit (where a Gaussian and a Lorentzian are folded), yields a correlation length of about 34 lattice cells along $\boldsymbol{c}$ (or 205 spins). Looking at the other widths in the critical regime, peaks can be still distinguished when the correlation length has fallen to about 120 spins, for example at 10 T and 10.76 K.

In the other figures Gauss fits are shown; the widths can in general be ascribed to the instrumental resolution, except in the case of the new phase, for which the (Gaussian) peaks are about 30% broader. At present the reason for this is not quite clear; domain formation could provide an explanation.

3. The magnetic structure factor

We want to illustrate how to read off from the diffraction peaks in figures 4–7 what the different structures may be, and how the spirals distort and evolve in the external field.

In zero field, the magnetic spirals are described by

$$\boldsymbol{S}_i(\boldsymbol{l}) = S\big(\cos(2\pi\boldsymbol{Q}\cdot\boldsymbol{l}+\phi_i), \sin(2\pi\boldsymbol{Q}\cdot\boldsymbol{l}+\phi_i), 0\big) \tag{3}$$

with

$$\phi_1 = 0 \qquad \phi_2 = 2\pi/3 \qquad \phi_3 = 4\pi/3$$
$$\boldsymbol{Q} = (0\,0\,Q).$$

We will use the chemical unit cell as the basis, so $Q = 6/L$, in units of c^*, where L is the (approximate) number of spins along the magnetic cell length.

For spirals distorted by the field such that higher harmonics are observed, the first step would be to expand a more general form than (3), as suggested by Jacobs *et al* (1993), for small fields:

$$S_{x1}(l) = S\cos\big(2\pi\boldsymbol{Q}\cdot\boldsymbol{l} - \delta\sin(2\pi\boldsymbol{Q}\cdot\boldsymbol{l})\big) \approx S\left(\cos(2\pi\boldsymbol{Q}\cdot\boldsymbol{l}) - \frac{\delta}{2}\cos(4\pi\boldsymbol{Q}\cdot\boldsymbol{l}) + \frac{\delta}{2}\right) \tag{4}$$

and similarly for the other components.

Even more generally, one can use the *ansatz*, with the field in the x-direction,

$$\begin{aligned} S_{xi}(l) &= a\cos(2\pi\boldsymbol{Q}\cdot\boldsymbol{l}+\phi_i) + b\cos(4\pi\boldsymbol{Q}\cdot\boldsymbol{l}+2\phi_i) + c \\ S_{yi}(l) &= a\sin(2\pi\boldsymbol{Q}\cdot\boldsymbol{l}+\phi_i) + b\sin(4\pi\boldsymbol{Q}\cdot\boldsymbol{l}+2\phi_i) \end{aligned} \tag{5}$$

with $3c = h$ from (2) with

$$h = \frac{g\mu_B H_x}{18SJ_1} \equiv \frac{H_x}{H_S}$$

and $H_S \approx 30$ T, the field at which saturation magnetization is found at low temperatures.

This *ansatz* was formulated with a kind of Landau theory in mind and involves the possibility of strong amplitude variations along the c-axis which should be admissible near the critical temperature as a result of fluctuations.

Up to constant factors, the magnetic scattering intensity is given by

$$F^2 = \sum_{lm}\sum_{ij}\sum_{\alpha\beta}(\delta_{\alpha\beta} - \tilde{k}_\alpha\tilde{k}_\beta)S_{li}^\alpha S_{mj}^\beta \exp\{2\pi\mathrm{i}\boldsymbol{k}\cdot(\boldsymbol{l}+\boldsymbol{d}_i-\boldsymbol{m}-\boldsymbol{d}_j)\} \tag{6}$$

where $\boldsymbol{k}$ is the scattering vector ($\tilde{\boldsymbol{k}}$ is normalized to unity), α and β denote Cartesian coordinates, $\boldsymbol{l}$ and $\boldsymbol{m}$ stand for lattice cell vectors along the c-axis and

$$\boldsymbol{d}_1 = (0\,0\,0) \qquad \boldsymbol{d}_2 = (1\,0\,0) \qquad \boldsymbol{d}_3 = 2\boldsymbol{d}_2 \tag{7}$$

for the three magnetic sublattices.

Introducing a shorthand notation for the Fourier transforms:

$$\frac{1}{N}\sum_{l} S^{x}_{li}\mathrm{e}^{2\pi\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{l}} = F\,[Sxi]$$

where N is the number of unit cells and i is the sublattice index, one obviously has to deal with terms of the type

$$\sum_{ij} F\,[Sxi]F^{*}\left[Sxj\right]\mathrm{e}^{2\pi\mathrm{i}\boldsymbol{k}\cdot(\boldsymbol{d}_i-\boldsymbol{d}_j)}$$

and similarly for the y-components, but no mixed x, y-terms. Averaging over left- and right-turning spirals, one ends up with the expression

$$I_x = \sum_{i} F\,[Sxi]F^{*}\,[Sxi] + \cos(2\pi\tilde{\boldsymbol{k}}\cdot\Delta\boldsymbol{d})\sum_{i\neq j} F\,[Sxi]\,F^{*}\left[Sxj\right] \tag{8}$$

and similarly for the y-part, $\Delta\boldsymbol{d}$ referring to any difference of the d_i-vectors. For peaks at reciprocal-lattice points of the type $(h\,h\,l)$, $\cos(2\pi\tilde{\boldsymbol{k}}\cdot\Delta\boldsymbol{d}) = 1$. We will not discuss these magnetic contributions to nuclear peaks further, since they turned out to be unmeasurable.

For peaks at reciprocal-lattice points of the type $(h \pm 1/3\,h \pm 1/3\,l)$, with l arbitrary, one has $\cos(2\pi\tilde{\boldsymbol{k}}\cdot\Delta\boldsymbol{d}) = -0.5$ and, for example, with (4) and (6),

$$F\,[Sxi] = \frac{h}{3}\delta(\boldsymbol{k}-\boldsymbol{\tau}) + \frac{a}{2}\sum_{\pm}\mathrm{e}^{\mp\mathrm{i}\phi_i}\delta(\boldsymbol{k}\pm\boldsymbol{Q}-\boldsymbol{\tau}) + \frac{b}{2}\sum_{\pm}\mathrm{e}^{\mp 2\mathrm{i}\phi_i}\delta(\boldsymbol{k}\pm 2\boldsymbol{Q}-\boldsymbol{\tau}) \tag{9}$$

where $\boldsymbol{\tau}$ is a reciprocal-lattice vector. It requires only a simple calculation to arrive at

$$I_x = \frac{9}{8}a^2\delta(1) + \frac{9}{8}b^2\delta(2) \tag{10}$$

with

$$\delta(1) = \delta(\boldsymbol{k}+\boldsymbol{Q}-\boldsymbol{\tau}) + \delta(\boldsymbol{k}-\boldsymbol{Q}-\boldsymbol{\tau})$$

and

$$\delta(2) = \delta(\boldsymbol{k}+2\boldsymbol{Q}-\boldsymbol{\tau}) + \delta(\boldsymbol{k}-2\boldsymbol{Q}-\boldsymbol{\tau}).$$

One obtains the same expression for the y-part.

Thus one can read off from the experiment, like in figure 4, how much the spirals are distorted. Experimentally, $a^2/b^2 \approx 3.5$ near the transition to the new phase; such a spiral, produced using (5), is shown in figure 8, with $Q = 0.052$. Of course, the parameter c remains experimentally invisible but could be guessed to be near 0.1, taking $h = 1/3$ near 10 T. This parameter allows for possible amplitude variations also in the IC1 phase.

The turning sense of the spirals is fixed by the DM interaction (in rare earths, for which spirals come about as a result of competing interactions along the c-axis, this is not so; see the book by Jensen and Mackintosh (1991)); it seems that, when the applied field forces the turning sense to change substantially, or the spins directed oppositely to the field are forced to spread too much, a phase transition is imminent—and is obviously possible for small spins via thermal fluctuations; then a softening effect can occur and, instead of the ‘wrong’ spins being spread, their amplitude becomes much smaller on the side away from the field; see figures 9 and 10.

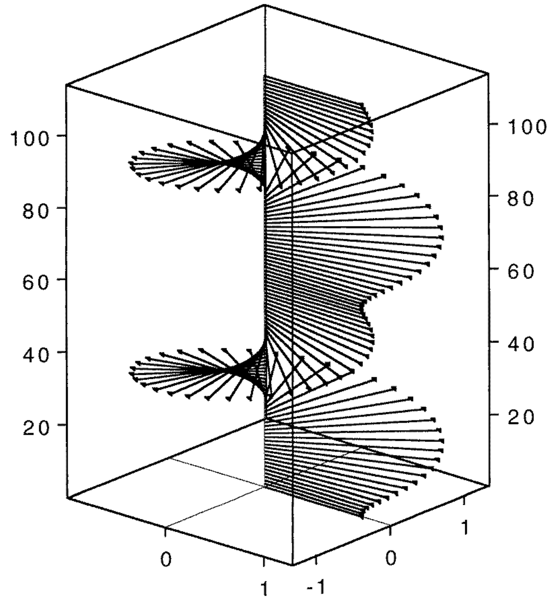

Figure 8. An illustration of a distorted spiral of the IC1 phase; the spirals of the two other sublattices look the same, but they are shifted by a third of their repeat length with respect to each other. The field direction can be deduced from the switch to the wrong turning sense. The parameters used here come in part from the experiment—see the text; here, explicitly, $S_x = \cos(2\pi l/115) - 0.53\cos(4\pi l/115) + 0.53$, and $S_y = \sin(2\pi l/115) - 0.53\sin(4\pi l/115)$.

Had the constant terms in (5) not been the same for all of the sublattices but—say—c_i, then a central component proportional to

$$\sum_i c_i^2 - \sum_{j>i} c_i c_j$$

would have resulted.

This indicates that a central component does not necessarily involve a commensurate structure. In fact, the experimental results are quite similar to those for the $\boldsymbol{H} \parallel \boldsymbol{c}$ case. There, the central component arose because there was a flop-like transition to the 'collinear' structure with two sublattice spins parallel. Here, this collinear structure is improbable, since—always assuming that the spins remain in the a–b plane—the angle of the spins with respect to the fields and each other would be uniquely determined by the field, and spiralling along $\boldsymbol{c}$ would contradict this. Also, it makes no sense to assume that the c_i should differ for the otherwise equivalent sublattices. Therefore we take up the structure proposal of Jacobs and Nikuni—see the following article—who assume two sublattices behaving very similarly, only with a phase shift of π—so they are also in a sense 'collinear', but antiparallel—and a third behaving distinctly differently, with the repeat length halved and a smaller amplitude variation. Illustrations of these two types of behaviour are shown in figures 9 and 10. It is interesting that the Landau theory yields optimal states with the expected larger $\boldsymbol{Q}_{\text{new}}$.

The order parameters of the new phase have the forms

$$\begin{aligned} S_{x1}(l) &= a\cos(2\pi \boldsymbol{Q}_{\text{new}} \cdot \boldsymbol{l}) - b\cos(4\pi \boldsymbol{Q}_{\text{new}} \cdot \boldsymbol{l}) + c \\ S_{x2}(l) &= -a\cos(2\pi \boldsymbol{Q}_{\text{new}} \cdot \boldsymbol{l}) - b\cos(4\pi \boldsymbol{Q}_{\text{new}} \cdot \boldsymbol{l}) + c \qquad (11a) \\ S_{x3}(l) &= 2b\cos(4\pi \boldsymbol{Q}_{\text{new}} \cdot \boldsymbol{l}) + d \end{aligned}$$

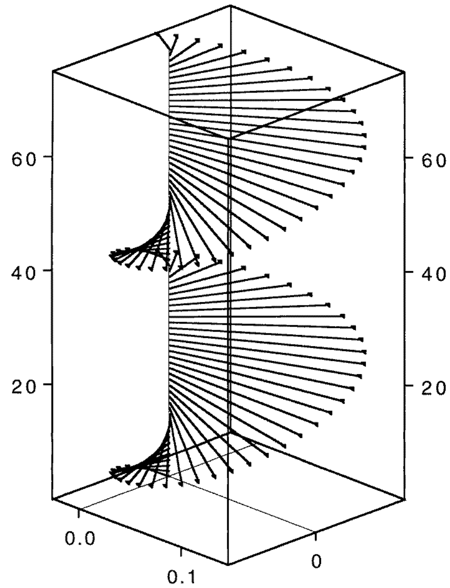

Figure 9. The 'spiral' of the new (IC2) phase; note the steady turning sense, however much the amplitudes are fluctuating. The field is towards the front right. As the field gets stronger some spins must pass through a 'zero-amplitude' state; this softening effect is thought to be due to thermal fluctuations near T_N. Here, explicitly, $S_x = -0.087\cos(2\pi l/76) - 0.009\cos(4\pi l/76) + 0.069$ and $S_y = -0.087\sin(2\pi l/76) - 0.009\sin(4\pi l/76)$. The other related sublattice with the phase shift of π would be described in the same way but with the minus signs changed to plus signs.

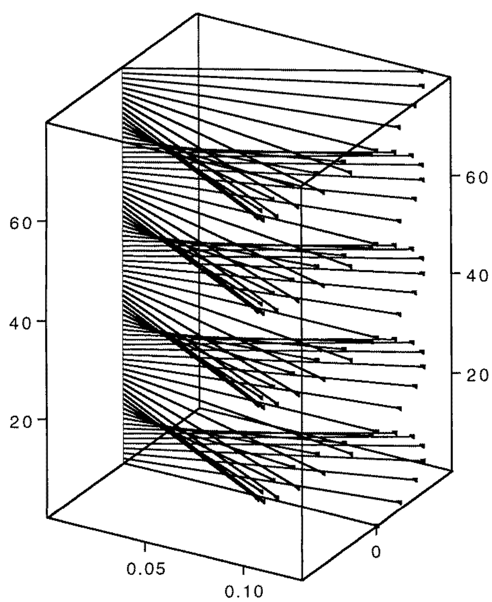

Figure 10. The third sublattice of the IC2 phase; according to figure 9 and (2), it must be described by $S_{x3} = 0.018\cos(4\pi l/76) + (h - 2 \times 0.069)$ and $S_{y3} = 0.018\sin(4\pi l/76)$, here drawn with $h = 0.25$. Most of the parameters cannot be read off from the experiments, as explained in the text.

$$
\begin{aligned}
S_{y1}(l) &= a\sin(2\pi \boldsymbol{Q}_{\text{new}}\cdot \boldsymbol{l}) - b\sin(4\pi \boldsymbol{Q}_{\text{new}}\cdot \boldsymbol{l})\\
S_{y2}(l) &= -a\sin(2\pi \boldsymbol{Q}_{\text{new}}\cdot \boldsymbol{l}) - b\sin(4\pi \boldsymbol{Q}_{\text{new}}\cdot \boldsymbol{l})\\
S_{y3}(l) &= 2b\sin(2\pi \boldsymbol{Q}_{\text{new}}\cdot \boldsymbol{l})
\end{aligned}
\tag{11b}
$$

with b much smaller than a and again $2c + d = h$.

The contributions to the magnetic structure factor can now easily be calculated as described above to give

$$
\begin{aligned}
F^2 &= F_x^2 + F_y^2\\
&= (1-\tilde{k}_x^2)\left((c-d)^2\delta(0) + \frac{3}{4}a^2\delta(1) + \frac{9}{4}b^2\delta(2)\right)\\
&\quad + (1-\tilde{k}_y^2)\left(\frac{3}{4}a^2\delta(1) + \frac{9}{4}b^2\delta(2)\right)\\
&= (1-\tilde{k}_x^2)(c-d)^2\delta(0) + (1+\tilde{k}_z^2)\left(\frac{3}{4}a^2\delta(1) + \frac{9}{4}b^2\delta(2)\right)
\end{aligned}
\tag{12}
$$

with $\delta(0) = \delta(\boldsymbol{k} - \boldsymbol{\tau})$.

According to the scattering geometry, the scattering vector is orthogonal to the field. With the choice of axes with $\boldsymbol{H}$ along $\boldsymbol{x}$, one has $\tilde{k}_x^2 \approx 0$ and $\tilde{k}_z^2 \approx 0$ near the reflections with $l = \pm Q$. For reflections with $l = 6 \pm Q$, one has $\tilde{k}_z^2 \approx 1$, so the relative weight of the peaks should then be different. Since the present results have exclusively been collected near $(\frac{1}{3}\,\frac{1}{3}\,Q)$, the peak ratio is given by $(c-d)^2/(3a^2/4)$, which, experimentally, is near 1/2. For figures 9 and 10, this ratio is nearer to 1/3 with the parameters provided from the Landau theory. Note that the calculations involve optimization with respect to many more parameters than the experiment provides: it provides only Q and the peak ratio, which already 'hides' three. Also, the third sublattice with vector $2\boldsymbol{Q}_{\text{new}}$ remains totally invisible in the experiment; it should appear in the next-higher harmonics of the side peaks of figure 6, which have not been observed—or, rather, were not looked for.

There is probably a chance of an observation when collecting data near $(1/3\;1/3\;6 \pm Q)$ with a wider l-scan range.

It seems plausible that the transition to the new phase is of first order, because the two phases are symmetrically unrelated, and that the transition to paramagnetic saturation is of second order, also because of the long correlation length. The 'spirals' no longer necessarily spiral around the c-axis, and in higher fields they will all move towards the field direction, the 'wrong' spins passing through a 'zero-amplitude' state. Thus there is a way for the spins to follow the field without stretching out the spiral, namely by means of a 'softening' of the amplitudes.

Beyond T_N, the peaks disappear into critical scattering for—within experimental accuracy—an unchanged spiral vector $\boldsymbol{Q}_{\text{new}}$, like for an ordinary antiferromagnet with sublattice magnetization as the order parameter.

4. Summary

Two incommensurate structures of $CsCuCl_3$ in an external field orthogonal to the hexagonal c-axis have been identified.

The first is the IC1 phase, which is a structure with three equivalent sublattices of distorted spirals, distinguished by a phase shift of $2\pi/3$ along $\boldsymbol{c}$, for which the neutron diffraction gives peaks at $(h \pm 1/3\; h \pm 1/3 \pm Q)$ and their second harmonics at

$(h \pm 1/3\, h \pm 1/3 \pm 2Q)$. We found that Q drops considerably in the field, but we never found it to reach zero, which means that in our measurements in the vicinity of the critical temperature we could not find a commensurate phase separating the incommensurate phase from the paramagnetic one.

The other phase, IC2, is characterized by diffraction peaks with a central component at $(h \pm 1/3\, h \pm 1/3\, 0)$ and side peaks of double the size at $(h \pm 1/3\, h \pm 1/3 \pm Q_{\mathrm{new}})$ with $Q_{\mathrm{new}} > Q$. The behaviour as the temperature approaches the critical value is also different: while for the IC1 phase the peaks stay sharp and Q falls, for the IC2 phase the peaks broaden and Q_{new} stays constant within experimental accuracy.

The new phase is extremely narrow, a few tenths of a degree Kelvin wide, lying croissant-like along the border between the IC1 and the paramagnetic phases.

The peaks of the new phase cannot be explained on the basis of a 120° structure of three equivalent sublattices, and an explanation is suggested by Jacobs and Nikuni (following paper) in terms of a structure with two sublattices behaving similarly—i.e. they are the same up to a phase shift of π—and differently from the third, which has a pitch of $2\boldsymbol{Q}_{\mathrm{new}}$

This structure agrees with the observations; however, since only the peak ratios and positions of the peaks are available as information from the experiment, the structure determination is not unique, as usual.

The phase transitions appear to be first order from IC1 to IC2 and second order from IC2 to the paramagnetic phase, in view of the presence of a two-phase regime for the first and critical scattering for the second. However, from the temperature dependence of the diffraction peaks and the field dependence of $\boldsymbol{Q}$, no reliable information as regards the order of the transitions and the critical exponents can be extracted.

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