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

A E Jacobs† and Tetsuro Nikuni‡

Department of Physics, University of Toronto, Toronto, Ontario, Canada M5S 1A7

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**Abstract.** $CsCuCl_3$ is a quantum triangular antiferromagnet, ferromagnetically stacked, with an incommensurate (IC) structure due to a Dzyaloshinskii–Moriya interaction. Because of the classical degeneracy caused by the frustration, fluctuations in $CsCuCl_3$ have extraordinarily large effects, such as the phase transition in longitudinal magnetic field (normal to the planes, parallel to the IC wavenumber $\boldsymbol{q}$) and the plateau in $\boldsymbol{q}$ in transverse field (perpendicular to $\boldsymbol{q}$). We argue that fluctuations are responsible also for the new IC phase discovered in transverse field near the Néel temperature $T_N$, by Werner *et al* (Werner T, Weber H B, Wosnitza J, Kelnberger A, Meschke M, Schotte U, Stüßer N, Ding Y and Winkelmann M 1997 *Solid State Commun.* **102** 609). We develop and analyse the corresponding minimal Landau theory; the ground-state reconstruction due to fluctuations is described phenomenologically, by means of a biquadratic term. The Landau theory gives two IC phases, one familiar from previous studies; the properties of the new IC phase, which occupies a pocket of the temperature–field phase diagram near $T_N$, agree qualitatively with those of the new phase found experimentally.

## 1. Introduction

Compounds based on the triangular antiferromagnet (TAFM), particularly members of the $ABX_3$ family, have provided a wealth of interesting behaviour and indeed many surprises, as recently reviewed [1]. Because of the extraordinarily large effects of quantum and thermal fluctuations, $CsCuCl_3$ (with Néel temperature [2] $T_N = 10.65$ K and zero-temperature saturation field $H_S \approx 30$ T) ranks among the most interesting of these compounds; the surprises began 20 years ago [3] and continue still, in theory as well as in experiment.

The magnetic properties of $CsCuCl_3$ are due to the $Cu^{2+}$ ions ($S = 1/2$); these form a stacked triangular lattice, to a good approximation. The interaction in the planes is antiferromagnetic and therefore frustrated. Normal to the planes (in the chain or $c$-direction), a ferromagnetic interaction competes with a Dzyaloshinskii–Moriya (DM) interaction [4], giving an incommensurate (IC) structure with wavenumber $\boldsymbol{q} = q\hat{\boldsymbol{z}}$ in the $c$-direction. In more detail, the classical, zero-temperature structure in zero magnetic field is the three-sublattice, $\pm 120^\circ$ TAFM structure; the spins lie in the planes, rotating by $\approx 5.1^\circ$ per plane [5]. Application of a magnetic field yields a variety of interesting phenomena related to the classical degeneracy of the TAFM; recall that the classical ground state of the TAFM is continuously (and also discretely) degenerate, even in a magnetic field, and also that thermal fluctuations [6–9] in classical models and quantum fluctuations [10–12] break the

† E-mail: jacobs@physics.utoronto.ca.

‡ Present address: Department of Physics, Tokyo Institute of Technology, Oh-okayama, Meguro-ku, Tokyo, Japan 152; e-mail: nick@stat.phys.titech.ac.jp.w.

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continuous degeneracy in the same way, both selecting for example the collinear structure at $H \approx H_S/3$.

In longitudinal magnetic field (normal to the planes, parallel to $\boldsymbol{q}$), the low-temperature magnetization is discontinuous [3, 13, 14] at $H \approx 0.4H_S$, due to a novel, fluctuation-induced phase transition [15, 16]: the umbrella structure is optimal at small $H$ (due to a small, easy-plane anisotropy in the interplane exchange [17]) and a coplanar structure is optimal at larger $H$ (due to quantum fluctuations). Other experiments [18–25] support the Nikuni–Shiba analysis [15, 16]. In summary, $CsCuCl_3$ in longitudinal field appears to be understood, except that the transition at high temperature $T$ has puzzling features [25].

A transverse field (in the planes, perpendicular to $\boldsymbol{q}$) gives more surprises. The behaviour at low fields is conventional and understood from classical (mean-field) theory [26, 27]: $q$ decreases quadratically with $H$, and the curvature increases [20, 22] with $T$. Classical theory [26] predicts a transition to the commensurate (C) phase at $H \approx 0.47H_S$ (at $T = 0$), as recently observed [28] (though at $H \approx 0.58H_S$). Classical theory fails in other respects. At low $T$, unusual behaviour occurs for fields near $H = H_S/3$, well below the IC $\rightarrow$ C transition; structure is found in the magnetization [14], in the $^{133}Cs$ NMR shift [23], in the IC wavenumber (reference [20] finds a plateau), and in ESR measurements [21]. At high $T$, a second IC phase appears, and $T_N$ increases with field [29].

The phase diagram near $T_N$ was recently found [30] for field orientations between longitudinal and transverse.

In transverse field, the structure near $H_S/3$ at low $T$ seems related to changes, induced by quantum fluctuations, in the structure of the TAFM near the same field [10–12]. Linear spin-wave (LSW) theory, which adds the leading quantum correction to classical theory, gives a plateau in the magnetization of the C state [31, 32] (as for the TAFM), a promising start. But there is another surprise, this time in theory. Not only does LSW theory of the IC phase fail to explain any of the other results, it actually provides a worse description of the IC phase than does classical theory, by predicting a premature IC $\rightarrow$ C transition [31, 32] at $H \approx 0.32H_S$. LSW theory fails because it omits the major effect of fluctuations, namely the reconstruction of the IC ground state, an effect apparently new with $CsCuCl_3$. An innovative, phenomenological method [31, 32] for treating fluctuations was proposed, justified (by verifying that it explains qualitatively the magnetization plateau in the TAFM), and then used to find the order parameter in the IC phase of $CsCuCl_3$ in transverse field. The wavenumber plateau [20, 28] near $H \approx H_S/3$ was successfully explained, even its level; the theoretical value ($H \approx 0.44H_S$) for the field at the IC $\rightarrow$ C transition [28] is however too small, and the magnetization is not predicted well, suggesting that the phenomenological theory can be improved.

Both new findings [29] near $T_N$, namely the increase of $T_N$ with field (as in the TAFM [6, 8, 33]) and the second IC phase, are probably due to thermal fluctuations; neither has yet been treated theoretically. Because a microscopic or numerical treatment of fluctuations is out of the question for a vector-spin system with a nonsinusoidal IC structure, we use phenomenology. To treat a particular aspect of fluctuations near $T_N$, namely the breaking of the classical TAFM degeneracy and the resulting thermal reconstruction of the IC ground state, we add to the standard Landau theory a term biquadratic in the order parameter, as in our treatment [31, 32] of quantum fluctuations at $T = 0$; this term appears neither in the Hamiltonian nor in mean-field theory [27] at any $T$. Of course this term is not intended to include fluctuation effects in general (such phenomenology cannot possibly explain the reduction of $T_N$ from the mean-field value of 35.5 K to the experimental value of 10.65 K).

In qualitative agreement with experiment [29], the Landau theory predicts a second IC phase to exist in a small $T$–$H$ region near $T_N$. As discussed in the preceding article [34]

and in section 4, the theory explains other properties of the new phase, qualitatively, though not the increase of $T_N$ with $H$. The new phase is the high-$T$ version of a state which arose in the classical theory [26]; the state exists at all $T$ in classical (mean-field) theory [26, 27], but is never optimal. The new phase owes its existence to thermal fluctuations; these are strong enough to overcome a small classical energy difference, just as quantum fluctuations in longitudinal field overcome the small anisotropy [15, 16]. A unifying feature is that the two most striking of the experimental results in transverse field, namely the plateau [20, 28] in $q$ and the new phase [29] near $T_N$, are explained using the same phenomenological treatment of fluctuations, in [31, 32] and here respectively.

Remarkably then, $CsCuCl_3$ displays a fluctuation-induced phase transition in transverse field, and a different fluctuation-induced phase transition in longitudinal field.

## 2. Hamiltonian

The main interactions are described by the Hamiltonian

$$\mathcal{H} = \sum_{in}\Big[-2J_0\boldsymbol{S}_{in}\cdot\boldsymbol{S}_{i,n+1} - D\hat{\boldsymbol{z}}\cdot(\boldsymbol{S}_{in}\times\boldsymbol{S}_{i,n+1}) + J_1\sum_{k}{}'\,\boldsymbol{S}_{in}\cdot\boldsymbol{S}_{kn} - g\mu_B H\hat{\boldsymbol{x}}\cdot\boldsymbol{S}_{in}\Big] \tag{1}$$

where $\boldsymbol{S}_{in}$ is the spin operator at the $i$th site in the $n$th $a$–$b$ plane, $\hat{\boldsymbol{z}}$ and $\hat{\boldsymbol{x}}$ are unit vectors in the $c$- and $a$-directions, and the $k$-sum is over the six, in-plane, nearest neighbours of the site $in$. The first term ($\propto J_0$) is the isotropic, ferromagnetic exchange interaction between spins in nearest-neighbour planes, the second ($\propto D$) is the interplane DM interaction, the third ($\propto J_1$) is the isotropic, frustrated, antiferromagnetic exchange interaction between nearest-neighbour spins in the $a$–$b$ planes, and the fourth is the Zeeman energy in a field $\boldsymbol{H}$ transverse to the chains. We omit the easy-plane anisotropy [17] in the interplane interaction, the dipole–dipole interaction, and several other effects. The coefficients have been estimated previously [2, 5, 35, 36, 17, 37]; we use $J_0 = 28$ K, $J_1 = 4.9$ K, and $D = 5$ K. The saturation field, above which each spin is aligned with the field at $T = 0$, is $H_S = 18J_1S/(g\mu_B) \approx 30$ T.

At the classical level, the intrachain exchange term ($J_0$) favours states with spins parallel in adjacent $a$–$b$ planes while the smaller DM term ($D$) favours states with spins in the planes and rotating by $\pi/2$ per plane. At zero field, for all $T < T_N$, the spins lie in the planes, forming the 120° structure with three sublattices. The structure normal to the planes is helical; the wavenumber at $H = 0$ is $q_0\hat{\boldsymbol{z}}$ where $q_0 = \arctan(D/(2J_0)) \approx 2\pi/71$. A transverse field deforms the helical structure, which becomes highly nonsinusoidal at higher $H$.

For $T = 0$, the above Hamiltonian was investigated in the classical approximation [26], and the leading quantum correction was obtained using LSW theory [31, 32]. As discussed above, neither theory can account for the structure observed near $H_S/3$.

The extension of classical theory to $T > 0$ (by mean-field theory) gives a phase diagram [27] with only one IC phase, and so an understanding of the new IC phase near $T_N$ seems to require including fluctuations at some level. A satisfactory microscopic treatment of fluctuations in $CsCuCl_3$ is out of the question at $T = 0$, and they are even more difficult to treat for $T > 0$, leaving it seems only a phenomenological approach. For general $T > 0$, one could simply add the biquadratic term [31, 32] to the mean-field expression [27] for the free energy, but the coefficient would have to be adjusted as fluctuation effects increase with $T$, requiring a fit at each $T$ or strong guidance from theory; this approach would be

most reasonable near $T_N$, if the mean-field free energy were expanded to fourth order in the order parameter. We have chosen instead to use a fourth-order Landau theory.

## 3. Landau theory

The following describes a minimal Landau theory of $CsCuCl_3$ near $T_N$; usually, Landau theory is reliable regarding the phase diagram, less reliable regarding the order of the transitions, and unreliable regarding fine details like the position dependence of the order parameter. We assume a structure with three sublattices in the $a$–$b$ planes and with period $L$ in the $c$-direction (in units of the layer spacing); the restriction to integer $L$ causes no difficulty [32]. We assume also that the spins remain in the $a$–$b$ planes at all $H$ and $T$. Curiously, the DM term is not sufficient for this [38, 32]; the easy-plane anisotropy [17] helps of course, but fluctuation effects seem to be necessary [32]. In mean-field theory, the free energy would be expressed in terms of the site-dependent magnetization $\langle \boldsymbol{S}_{jl} \rangle$, where $j = 1, 2, 3$ is the sublattice index and $l = 1, \ldots, L$ is the layer index. In Landau theory, the free energy is expanded in the order parameter $\boldsymbol{m}_{jl}$, which is only proportional to $\langle \boldsymbol{S}_{jl} \rangle$.

Explicitly, we use the following expression for the free energy $F$ of the $N$ spins, relative to the paramagnetic state at $H = 0$:

$$F = \frac{N}{3L} \sum_{j=1}^{3} \sum_{l=1}^{L} \Big[ \tfrac{1}{2}\alpha_1 \boldsymbol{m}_{jl}^2 + \alpha_2 \boldsymbol{m}_{jl} \cdot \boldsymbol{m}_{j+1,l} - h\hat{\boldsymbol{x}} \cdot \boldsymbol{m}_{jl} + \tfrac{1}{4}\gamma_1 \boldsymbol{m}_{jl}^4 - \tfrac{1}{2}\gamma_2 (\boldsymbol{m}_{jl} \cdot \boldsymbol{m}_{j+1,l})^2 - \delta_1 \boldsymbol{m}_{jl} \cdot (\boldsymbol{m}_{j,l+1} - \boldsymbol{m}_{jl}) - \delta_2 \hat{\boldsymbol{z}} \cdot (\boldsymbol{m}_{jl} \times \boldsymbol{m}_{j,l+1}) \Big]. \tag{2}$$

For stability, $\gamma_1 > 0$ and $\gamma_2 < 0.5\gamma_1$. This expression differs in major respects from the mean-field expansion to fourth order in $\boldsymbol{m}$ (or $\langle \boldsymbol{S} \rangle$). First, we omit many terms of the same order as the ones that we keep, second order as well as fourth order; more importantly, we add a fourth-order term (coefficient $\gamma_2$) which does not appear in the expansion, as discussed in the next paragraph. All seven terms are essential. The terms with coefficients $\alpha_1$ and $\gamma_1$ are standard, while those with coefficients $\alpha_2$, $\delta_1$, and $\delta_2$ result respectively from the in-plane antiferromagnetic interaction, the interplane ferromagnetic interaction, and the DM interaction; the terms in $\alpha_1$ and $\delta_1$ are adjusted so that the latter makes no contribution to the energy of the commensurate state. The term with Landau parameter $h$ (proportional to the magnetic field $H$) is the Zeeman energy.

The remaining term, the biquadratic term $-\frac{1}{2}\gamma_2(\boldsymbol{m}_{jl} \cdot \boldsymbol{m}_{j+1,l})^2$, appears neither in the Hamiltonian nor in the mean-field theory [27]; it represents some effects of thermal fluctuations, at the phenomenological level [39]. In the TAFM and in the C state of $CsCuCl_3$ [26], fluctuations break the classical degeneracy, selecting one state at the LSW level. Their effect on the IC phase of $CsCuCl_3$ is more profound. First, the IC structure forbids the selection possible in a C structure. Second, the classical IC phase is unconventional, being well described as a continuous sequence of degenerate C states [26]. Fluctuations break the degeneracy, reorienting the spins to give a conventional IC phase with domain walls separating nearly C regions. LSW theory of the IC phase fails because it cannot account for the reorientation and the resulting reconstruction of the IC ground state.

From the Hamiltonian, we expect the coefficients $\alpha_2$, $\delta_1$, and $\delta_2$ to be proportional to $6J_1$, $2J_0$, and $D$ respectively. To reduce the number of parameters, we normalize the order parameter and the free energy so that $\alpha_2 = 1$ and $\gamma_1 = 1$. Then the interlayer coefficients are $\delta_1 = 2J_0/(6J_1) \approx 1.9$ and $\delta_2 = D/(6J_1) \approx 0.17$. Of the remaining parameters $\alpha_1$, $h$, and $\gamma_2$, only $\alpha_1$ depends on $T$. At $H = 0$, the IC–paramagnetic transition at $T_N(0)$ occurs at $\alpha_1 \approx 1.015$; $\alpha_1 = 1$ is the upper limit of the C phase at $H = 0$. In Landau theory, the

transition at $T_N(0)$ is second order. Experiment [25] finds tricritical or weakly first-order behaviour; the latter is obtained in a recent Monte Carlo analysis [51] of a related model.

The unknown constants of proportionality involving $\boldsymbol{m}$ and $h$ can be determined by comparing the Landau and mean-field theories. From equation (2), the Landau energy (per spin) of the paramagnetic state at $\alpha_1 = 1$ is $\frac{3}{2}m^2 - hm + \frac{1}{4}m^4$. The corresponding mean-field expression is found by setting $\beta S^2 J_C = 1$ in equation (3.2) of [27]:

$$\frac{\mathcal{J}}{J_C}\left(18J_1\langle S\rangle^2 - 2g\mu_B H\langle S\rangle + \frac{4}{3S^2}\frac{\mathcal{J}^3}{J_C^2}\langle S\rangle^4 + \cdots\right)$$

where $J_C = 4J_0 + 6J_1$ and $\mathcal{J} = 2J_0 - 6J_1$. Because the mean-field expression omits fluctuations, the term in $\gamma_2$ must be omitted from the Landau expression. On setting $\langle S\rangle = am$ and comparing coefficients, one finds $h = kH/H_S(0)$ where $k = 3S/a$ and $k^2 = 4\mathcal{J}^3/(J_C^2 J_1)$; the numerical value is $k \approx 0.88$.

The Euler–Lagrange equations of the above Landau theory have many solutions, namely the paramagnetic (P) solution $\boldsymbol{m}_{jl} = m_P\hat{\boldsymbol{x}}$, several commensurate (C) solutions, and many incommensurate (IC) solutions; the following provides some background for the last. In classical theory [26], the magnitudes of the site magnetizations are fixed and the phases suffice for a complete description. A single IC solution is optimal at all fields below the IC $\rightarrow$ C transition (predicted to occur at $H \approx 0.47H_S$); in this solution (called the 111 solution), the spins on all three sublattices wind through $2\pi$ over a period $L$ of the IC structure. Many other IC solutions exist at fields below the transition. In the 110 solution, which is never optimal, the spins on only sublattices 1 and 2 wind through $2\pi$ over one period, while the spins on sublattice 3 wobble about the field without winding. The 111 and 110 solutions, and many other solutions generated from them by forming composite solutions [26], become degenerate at the IC $\rightarrow$ C transition, which is therefore a multiphase point. In mean-field theory [27], for $T > 0$, the magnitudes of the magnetizations are no longer fixed; they can adjust to minimize the energy (for example by decreasing near a domain wall). The solution corresponding to the 111 solution is again optimal in all cases; it loses its winding character at larger $T$ and $H$ where the orbit in the $m_x$–$m_y$ plane no longer encircles the origin. The infinite degeneracy at the IC $\rightarrow$ C transition remains at $T > 0$.

The Landau-theory states corresponding to the 111 and 110 solutions are most easily described at $H = 0$ (where the wavenumber is $q_0$ for both). For the first ($IC_1$),

$$\boldsymbol{m}_{jl} = m_1\left[\hat{\boldsymbol{x}}\cos(q_0 l + \phi_j) + \hat{\boldsymbol{y}}\sin(q_0 l + \phi_j)\right] \qquad \text{for } j = 1, 2, 3 \tag{3}$$

with $\phi_j = \phi_0 + (j-1)2\pi/3$. For the second ($IC_2$),

$$\boldsymbol{m}_{1l} = -\boldsymbol{m}_{2l} = m_2\left[\hat{\boldsymbol{x}}\cos(q_0 l + \phi_0) + \hat{\boldsymbol{y}}\sin(q_0 l + \phi_0)\right] \qquad \boldsymbol{m}_{3l} = 0. \tag{4}$$

This is stable at $H = 0$ only for $\alpha_1 \gtrsim 0.99$, but solutions at lower $\alpha_1$ and $H > 0$ are easily found. The first has the lower energy at $H = 0$, for all $T$. For the same amplitude, the first optimizes the $\alpha_2$-term in the density of equation (2), while the second optimizes the biquadratic term; the respective energies are $-\frac{3}{2}\alpha_2 m^2 - \frac{3}{8}\gamma_2 m^4$ and $-\alpha_2 m^2 - \frac{1}{2}\gamma_2 m^4$. It is then possible that the second can have the lower energy, though only for $H > 0$.

For $H > 0$, some analytical results can be found at small field [26, 27], but full numerical solutions of the Euler–Lagrange equations are required at general values of $H$. Solutions were found by repeated linearization about trial solutions and solution of the linearized equations for the corrections. With increasing $H$, both solutions evolve, becoming increasingly nonsinusoidal; other solutions are found, but these are never optimal. In the second solution, the order parameter on the third sublattice increases from 0 and wobbles

about $\boldsymbol{H}$ with period $L/2$. Phase diagrams follow from comparison of the energies of the IC, P, and C solutions.

## 4. The phase diagram and other results

Because equation (2) cannot explain both the new IC phase and the increase of $T_N$ with $H$, and also because equation (2) omits many terms of the same order as those kept, a detailed comparison with experiment is not attempted; fine details of the results should not be relied upon. The only parameter in the theory (apart from the unknown constant relating $\alpha_1$ to $T$) is the fluctuation coefficient $\gamma_2$; its sign is positive [31, 32], but the strength of the fluctuations is unknown, and so its magnitude is adjustable. The value $\gamma_2 = 0.2$ used in the following was chosen, with guidance from experiment [34], to give a reasonable size to the $IC_2$ region of the phase diagram, and also to the plateau in $q/q_0$ for the $IC_1$ phase (as a function of $h$); 0.1 seems too small and 0.3 too large. Actually, the new phase appears even at $\gamma_2 = 0$, but only in a thin sliver ($\Delta h \leqslant 0.024$) of the phase diagram, with re-entrance and with no sizable plateau.

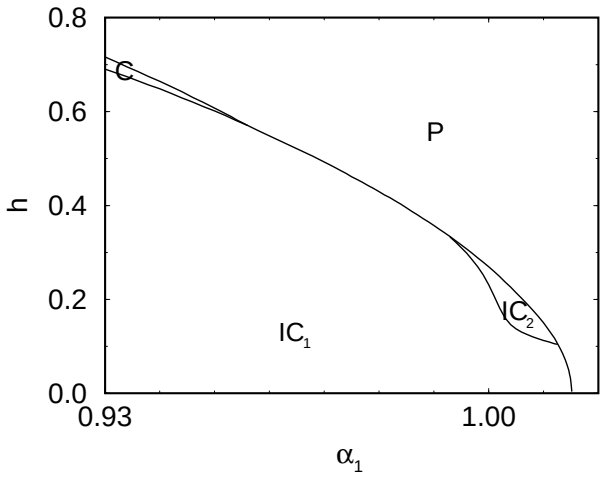


**Figure 1.** The phase diagram in the $\alpha_1$–$h$ plane for $\gamma_2 = 0.2$. The Landau parameter $\alpha_1$ is linear in the temperature $T$, and $h \approx 0.88H/H_S(0)$. The paramagnetic phase (P), the commensurate phase (C), and the two incommensurate phases ($IC_1$ and $IC_2$) are optimal in the regions indicated.

### *4.1. The new phase*

Figure 1 shows the theoretical phase diagram near $T_N$; again, $\alpha_1$ is linear in $T$ ($\alpha_1 = 1.015$ at $T_N(H = 0)$) and $h \approx 0.88H/H_S(0)$. The two IC phases ($IC_1$ and $IC_2$), the P phase, and the C phase are optimal in the regions indicated; the C phase is the $\boldsymbol{m}_1 = \boldsymbol{m}_2$ phase of the stacked TAFM. The new feature here is the $IC_2$ phase.

The companion article [34] presents the strongest evidence for identifying the $IC_2$ phase with the new IC phase discovered in [29]: the neutron-scattering intensity as a function of wavenumber is qualitatively that expected from the order parameter of the $IC_2$ phase. Reference [34] compares theory and experiment in other respects as well. The following compares several aspects of the phase diagrams; it also presents results for the dependence of the IC wavenumbers on $T$ and $H$, and for the order parameters in the two IC phases.

### *4.2. The phase diagram (qualitative aspects)*

In figure 1, as in figure 3 of [34], the new phase appears in a small $T$–$H$ region near $T_N$. In both, the new phase appears only above some field, and a narrow tail extends to the low-$T$ side. Landau theory misses the increase of $T_N$ with $H$ and therefore also the nose; experimentally, the region of the new phase is shaped more like a croissant than a pocket.

### *4.3. The phase diagram (the order of the transitions)*

The fields available in the high-$T$ neutron measurements [29, 34] were not sufficient to observe the C phase, nor of course the $IC_1 \rightarrow C$ and $C \rightarrow P$ transitions; the $IC_1 \rightarrow C$ transition was however observed [28] at low $T$, at $H \approx 18$ T.

In theory, all four of the other transitions in figure 1 are first order. $IC_1 \rightarrow IC_2$ is strongly first order, while the three transitions to the P state ($IC_1 \rightarrow P$ near $T_N$, $IC_2 \rightarrow P$ and $IC_1 \rightarrow P$ at lower $T$) are weakly first order (the free energies cross with almost the same slope); the $IC_1 \rightarrow P$ transition is second order at $H = 0$.

In experiment, only $IC_1 \rightarrow IC_2$ is unambiguously first order. The scan [34] at 10.34 K can be interpreted in two ways [52]: either the tail of the $IC_2$ phase was missed, or there is a first-order $IC_1 \rightarrow P$ transition at $H \approx 12$ T. The $IC_2 \rightarrow P$ transition is almost certainly second order, from the observation of critical scattering [34].

### *4.4. The phase diagram (other aspects)*

In theory, the $IC_2$ phase is not found at any $T$ if $h < 0.10$ or if $h > 0.34$, corresponding to $H < 3.4$ T and $H > 11.6$ T. In experiment, the lower limit is $H = 4.3 \pm 0.3$ T; if the $IC_2$ phase does not appear at 10.34 K, then the upper limit is $11.5 \pm 0.5$ T. The agreement is reasonable. Independent of the constant $k$ relating $h$ and $H$, the relative widths in the field variable are comparable: $11.5/4.3 \approx 0.34/0.10$. The nose in the experimental phase diagram prevents a similar analysis for the temperature variable; for example, we cannot estimate reliably the upper $T$-limit of the C phase (but none of our estimates disagrees with the data).

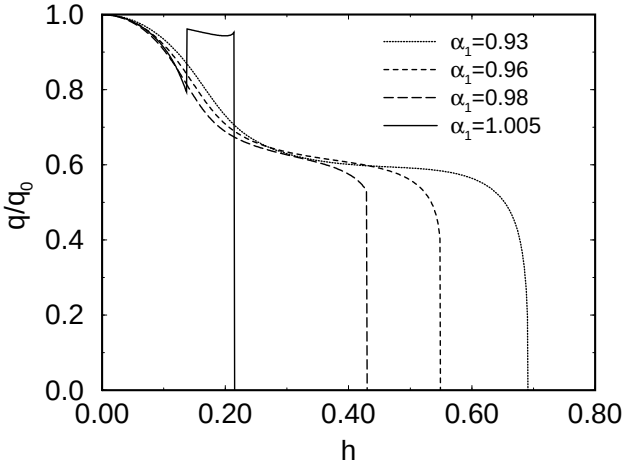


**Figure 2.** The reduced wavenumber $q/q_0$ as a function of the Landau field $h$ for four values of the Landau parameter $\alpha_1$.

### *4.5. Wavenumbers of the IC phases*

Figure 2 shows theoretical results for the reduced wavenumber $q/q_0$ as a function of $h$, at four values of $\alpha_1$.

For $\alpha_1 = 1.005$, there are two transitions as $h$ increases, $IC_1 \rightarrow IC_2$ at $h \approx 0.137$ and $IC_2 \rightarrow P$ at $h \approx 0.202$, both first order in theory; at the first, $q$ increases discontinuously to a value less than the zero-field value $q_0$. In theory, $q/q_0$ for the $IC_2$ phase is roughly independent of $h$, for fixed $\alpha_1$. The dependence on $\alpha_1$ is stronger, but still weak; the value decreases roughly linearly with $\alpha_1$, from $q/q_0 \approx 0.98$ at $\alpha_1 = 1.01$ to $q/q_0 \approx 0.88$ at $\alpha_1 = 0.995$. The experimental value [34] for $q/q_0$ in the $IC_2$ phase is $\approx$0.87; this is larger than in the $IC_1$ phase, as in theory; on the other hand, no dependence on $T$ or $H$ was observed.

At lower $T$ ($\alpha_1 = 0.98$ and 0.96), $q$ decreases as $h$ increases, flattens out, bends over, and then drops discontinuously in a first-order transition to the P phase. At $\alpha_1 = 0.93$ (apparently corresponding to lower $T$ than used in [34]), $q$ forms a reasonable plateau before rounding and falling to zero in a weakly first-order transition to the C phase; at slightly larger $h$, a second-order transition occurs to the P phase.

Theoretically, the plateau in $q/q_0$ occurs at $\approx$0.6, almost independently of $\alpha_1$; the level of the plateau is reasonably robust (for $\gamma_2 = 0.3$, the plateau occurs at $q/q_0 \approx 0.56$). Figure 1 of the preceding article [34] compares these results with the available data. Experiment finds a plateau (as in theory), at about the theoretical level ($q/q_0 \approx 0.6$), for both $T = 10.34$ K and $T = 9.95$ K; the latter data are slightly rounded at higher $H$, as in theory for $\alpha_1 \lesssim 0.98$. Data were not obtained at low fields where $q$ descends from the zero-field value to the plateau, preventing more detailed comparison with theory.

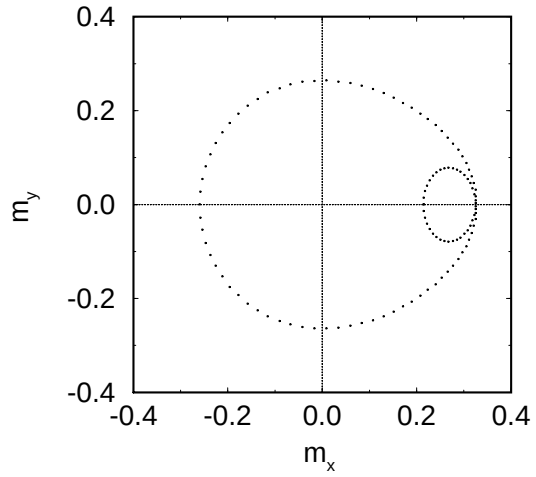


**Figure 3.** The $IC_1$ phase: the orbit in the $m_x$–$m_y$ plane for the order parameter on one of the three equivalent sublattices. The Landau parameters are $\alpha_1 = 0.96$ and $h = 0.4$ (near the middle of the plateau in figure 2); the period $L$ is 116. The order parameters on the other two sublattices are displaced by $l = \pm L/3$ from the first.

### *4.6. Order parameters*

Figure 3 shows the order parameter for the $IC_1$ phase, for Landau parameters $\alpha_1 = 0.96$ and $h = 0.4$ (near the middle of the plateau in figure 2). At maximum $m_x$, the configuration

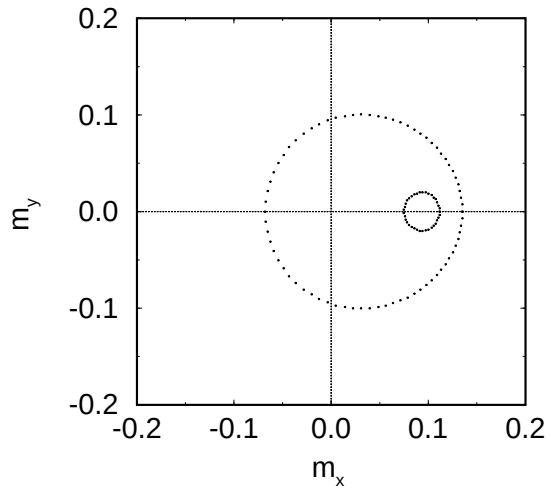


**Figure 4.** The $IC_2$ phase: the orbits in the $m_x$–$m_y$ plane for the order parameters on two of the three sublattices. The Landau parameters are $\alpha_1 = 1.005$ and $h = 0.18$ (near the middle of the pocket in figure 1); the period $L$ is 74. The outer loop is the orbit for one of the two equivalent sublattices; the orbit for the second is displaced by $l = L/2$. The order parameter on the third sublattice (inner loop) wobbles about the field with period $L/2$.

is almost collinear. Figure 4 shows the order parameter for the $IC_2$ phase, for $\alpha_1 = 1.005$ and $h = 0.18$ (near the middle of the pocket in figure 1). Figures 8 to 10 of [34] provide other views of the order parameters in the two IC phases.

Neutron scattering does not determine the order parameter uniquely, and so detailed comparison with experiment is not possible. As discussed in the companion article [34], Landau theory succeeds qualitatively in describing the experimental results, particularly the central component for the $IC_2$ phase. The theoretical order parameter in the $IC_1$ phase (figure 3) is too distorted, due to the retrograde motion. That in the $IC_2$ phase agrees reasonably well, although the third sublattice is not visible in the available data.

## 5. Summary

Landau theory with the biquadratic term explains the appearance of the new IC phase found [29, 34] near $T_N$. The new phase is the Landau-theory counterpart of the 110 state studied in [26], but stabilized by fluctuations. Only coarse adjustment of the only available parameter (the coefficient $\gamma_2$ of the biquadratic term) is needed to obtain qualitative (in some cases quantitative) agreement with experiment.

In more detail: Landau theory finds a new IC phase to exist near $T_N$. It explains [34] qualitatively the neutron-diffraction results for both IC phases. It does not explain the increase of $T_N$ with field, but it explains other features of the phase diagram. It predicts moderately well the order of the transitions. It predicts that the wavenumber $q$ of the $IC_2$ phase is larger than in the $IC_1$ phase, as observed; the experimental $T$-dependence of $q$ is however weaker than predicted. It predicts that the plateau in the wavenumber of the $IC_1$ phase occurs at $q/q_0 \approx 0.6$, as observed. Theory and experiment agree qualitatively with respect to the order parameters.

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## Appendix. Extended Landau theory

The Néel temperature $T_N$ increases initially with field for the TAFM because of thermal fluctuations [6, 8, 33]. It increases also for $CsCuCl_3$ [29, 34], for (one believes) the same reason. Since equation (2) cannot explain the increase, it is natural to ask whether an extended Landau theory can do so. To investigate this question, we add to the square brackets in equation (2) the following sum of fourth-order invariants (from a Landau-mean-field theory [50] of the ferromagnetically stacked TAFM with $XY$-spins):

$$\tfrac{1}{4}\gamma_3 \boldsymbol{m}_{jl}^2 \sum_{k=1}^{3} \boldsymbol{m}_{kl}^2 + \gamma_4 \boldsymbol{m}_{jl}^2 \sum_{k=1}^{3} \boldsymbol{m}_{kl}\cdot\boldsymbol{m}_{k+1,l} + \tfrac{1}{2}\gamma_5 \boldsymbol{m}_{jl}\cdot\boldsymbol{m}_{j+1,l} \sum_{k=1}^{3} \boldsymbol{m}_{kl}\cdot\boldsymbol{m}_{k+1,l} + \gamma_6 \boldsymbol{m}_{jl}^2 (\boldsymbol{m}_{j-1,l}\cdot\boldsymbol{m}_{j+1,l}). \tag{A1}$$

Our coefficients are related to those of [50] by

$$\begin{aligned} B_1 &= \gamma_1 - \tfrac{1}{2}\gamma_2 + 3\gamma_3 - 6\gamma_4 + \tfrac{3}{2}\gamma_5 - 2\gamma_6 \\ B_2 &= \gamma_1 - 2\gamma_2 + 4\gamma_6 \\ B_3 &= \gamma_1 - 2\gamma_2 + 3\gamma_3 + 12\gamma_4 + 6\gamma_5 - 4\gamma_6 \\ B_4 &= \gamma_1 - \tfrac{1}{2}\gamma_2 - 2\gamma_6 \\ B_5 &= \gamma_1 + \gamma_2 + 3\gamma_3 + 3\gamma_4 - 3\gamma_5 + \gamma_6 \\ B_6 &= \gamma_1 + \gamma_2 + \gamma_6. \end{aligned} \tag{A2}$$

Equation (3) of [50], with $B_i = 9T_N/5$ for $i = 1$ to 6, reduces to our equation (2) with the last three terms (coefficients $\gamma_2$, $\delta_1$, and $\delta_2$) omitted, although some effort is needed to see this. The collinear phase cannot be obtained in mean-field theory; it appears at intermediate fields if only $B_4$ is different from (less than) the other $B_i$, or if $B_2$ (or $B_3$) is different [50].

For the stacked TAFM, at $T$ sufficiently below $T_N$, the phase sequence with decreasing $H$ must be: P phase $\rightarrow$ C phase with $\boldsymbol{m}_1 = \boldsymbol{m}_2 \rightarrow$ collinear C phase $\rightarrow$ low-field C phase; a little analysis gives the requirements $\gamma_2 > 0$, $B_3 > 0$ and $B_5 > 0$. If the P $\rightarrow$ C transition is second order (as at $h = 0$, $\alpha_1 = \alpha_2$), we find that the phase boundary is given by

$$h^2 = \left(\frac{\alpha_2 - \alpha_1}{B_5}\right)\left[(\alpha_1 + 2\alpha_2) + \frac{B_3}{B_5}(\alpha_2 - \alpha_1)\right]^2. \tag{A3}$$

Since $B_5 > 0$, this theory cannot explain the increase of $T_N$ with $H$ for the stacked TAFM; therefore we believe that it cannot explain the corresponding increase for $CsCuCl_3$.

We also used the extended theory to determine several phase diagrams like figure 1, for several different sets of parameters. The C, P, and $IC_1$ phases always appear, as does the $IC_2$ phase (unless of course $\gamma_2$ is sufficiently negative). Generally, the more complicated free energies (those with more of the $\gamma_i$-parameters $\neq 0$) give more complicated phase diagrams (with for example re-entrant phases), but no new phases are found.

## References


[1] Collins M F and Petrenko O A 1997 *Can. J. Phys.* **75** 605
[2] Achiwa N 1969 *J. Phys. Soc. Japan* **27** 561
[3] Motokawa M 1978 *Ann. Mtg of the Physical Society of Japan (Sendai)* unpublished
[4] Dzyaloshinskii I 1958 *J. Phys. Chem. Solids* **4** 241
Moriya T 1960 *Phys. Rev.* **120** 91
[5] Adachi K, Achiwa N and Mekata M 1980 *J. Phys. Soc. Japan* **49** 545
[6] Lee D H, Joannopoulos J D, Negele J W and Landau D P 1984 *Phys. Rev. Lett.* **52** 433
Lee D H, Joannopoulos J D, Negele J W and Landau D P 1986 *Phys. Rev.* B **33** 450
[7] Kawamura H 1984 *J. Phys. Soc. Japan* **53** 2452
Kawamura H 1991 *J. Phys. Soc. Japan* **61** 1299
[8] Kawamura H and Miyashita S 1984 *J. Phys. Soc. Japan* **53** 4138
Kawamura H and Miyashita S 1985 *J. Phys. Soc. Japan* **54** 4530
Miyashita S and Kawamura H 1985 *J. Phys. Soc. Japan* **54** 3385
[9] Korshunov S E 1986 *J. Phys. C: Solid State Phys.* **19** 5927
[10] Nishimori H and Miyashita S 1986 *J. Phys. Soc. Japan* **55** 4448
[11] Golosov D I and Chubukov A V 1989 *Pis. Zh. Eksp. Teor. Fiz.* **50** 416 (Engl. Transl. 1990 *JETP Lett.* **50** 451)
[12] Chubukov A V and Golosov D I 1991 *J. Phys.: Condens. Matter* **3** 69
[13] Fedoseeva N V, Gekht R S, Velikanova T A and Balaev A D 1985 *Pis. Zh. Eksp. Teor. Fiz.* **41** 332 (Engl. Transl. 1985 *JETP Lett.* **41** 406)
Gekht R S, Fedoseeva N V, Dolina V A and Balaev A D 1989 *Phys. Status Solidi* b **155** 639
[14] Nojiri H, Tokunaga Y and Motokawa M 1988 *J. Physique Coll.* **49** C8 1459
[15] Shiba H and Nikuni T 1993 *Recent Advances in Magnetism of Transition Metal Compounds* ed A Kotani and N Suzuki (Singapore: World Scientific) p 372
[16] Nikuni T and Shiba H 1993 *J. Phys. Soc. Japan* **62** 3268
[17] Tanaka H, Schotte U and Schotte K-D 1992 *J. Phys. Soc. Japan* **61** 1344
[18] Ohta H, Imagawa S, Motokawa M and Tanaka H 1993 *J. Phys. Soc. Japan* **62** 3011
[19] Chiba M, Ajiro Y and Morimoto T 1994 *Physica* B **201** 200
[20] Mino M, Ubukata K, Bokui T, Arai M, Tanaka H and Motokawa M 1994 *Physica* B **201** 213
[21] Ohta H, Imagawa S, Motokawa M and Tanaka H 1994 *Physica* B **201** 208
[22] Schotte U, Stüßer N, Schotte K-D, Weinfurter N, Mayer H M and Winkelmann M 1994 *J. Phys.: Condens. Matter* **6** 10 105
[23] Chiba M, Ajiro Y and Morimoto T 1995 *J. Magn. Magn. Mater.* **140–144** 1673
[24] Motokawa M and Arai M 1995 *Physica* B **213+214** 1017
[25] Weber H B, Werner T, Wosnitza J, von Löhneysen H and Schotte U 1996 *Phys. Rev.* B **54** 15 924
[26] Jacobs A E, Nikuni T and Shiba H 1993 *J. Phys. Soc. Japan* **62** 4066
[27] Ohyama T and Jacobs A E 1995 *Phys. Rev.* B **52** 4389
[28] Nojiri H, Takahashi K, Fukuda T, Fujita M, Arai M and Motokawa M 1997 *Int. Conf. on Neutron Scattering (Toronto)*
[29] Werner T, Weber H B, Wosnitza J, Kelnberger A, Meschke M, Schotte U, Stüßer N, Ding Y and Winkelmann M 1997 *Solid State Commun.* **102** 609
[30] Wosnitza J, Pérez F, Weber H B, Werner T, von Löhneysen H and Schotte U 1998 *J. Magn. Magn. Mater.* **177–181** 177
[31] Nikuni T 1995 *PhD Thesis* Tokyo Institute of Technology
[32] Nikuni T and Jacobs A E 1998 *Phys. Rev.* B **57** 5205
[33] Miyashita S 1986 *J. Phys. Soc. Japan* **55** 3605
[34] Schotte U, Kelnberger A and Stüßer N 1998 *J. Phys.: Condens. Matter* **10** 6391
[35] Hyodo H, Iio K and Nagata K 1981 *J. Phys. Soc. Japan* **50** 1545
[36] Tazuke Y, Tanaka H, Iio K and Nagata K 1981 *J. Phys. Soc. Japan* **50** 3919
[37] Mekata M, Ajiro Y, Sugino T, Oohara A, Ohara K, Yasuda S, Oohara Y and Yoshizawa H 1995 *J. Magn. Magn. Mater.* **140–144** 1987
[38] Jensen J 1995 unpublished
[39] The biquadratic term was used in this manner first in [31, 32]; it has appeared in other contexts, however. For systems with $S > 1/2$, the term does not necessarily represent fluctuation effects if it appears in the Landau or mean-field energies, as in [40, 41]. It has appeared, even in $S = 1/2$ systems, in the microscopic analysis of effects due to quantum fluctuations, thermal fluctuations, dilution, and magnetoelastic coupling

[42–49], as noted also in [12]; there exists however no microscopic derivation of the term as representing fluctuation effects for an arbitrary spin configuration. In [42–49], the analysis was limited to the LSW level, and the ground state was either known beforehand or selected from known candidates; an unpublished manuscript [46] used the term phenomenologically to estimate wall energies above a known ground state, the coefficient being fitted to numerical LSW results. In contrast, in [31, 32] the term includes fluctuation effects beyond the LSW level, explicitly the reconstruction of the IC ground state. As discussed in the appendix, reference [50] on the stacked TAFM did not consider the term explicitly; it noted however that some effects of fluctuations (explicitly the collinear phase) can be mimicked by allowing some Landau coefficients to depart from the mean-field values, thereby intentionally breaking the classical TAFM degeneracy.

[40] Sakakibara T and Date M 1984 *J. Phys. Soc. Japan* **53** 3599
[41] Mailhot A, Plumer M L, Caillé A and Azaria P 1992 *Phys. Rev.* B **45** 10 399
[42] Shender E F 1982 *Zh. Eksp. Teor. Fiz.* **83** 326 (Engl. Transl. 1982 *Sov. Phys.–JETP* **56** 178)
[43] Long M W 1989 *J. Phys.: Condens. Matter* **1** 2857
[44] Henley C L 1989 *Phys. Rev. Lett.* **62** 2056
[45] Chandra P, Coleman P and Larkin A I 1990 *Phys. Rev. Lett.* **64** 88
[46] Larson B E and Henley C L 1990 unpublished
[47] Henley C L 1994 *Phys. Rev. Lett.* **73** 2788
[48] Yıldırım T, Harris A B and Shender E F 1996 *Phys. Rev.* B **53** 6455
[49] Shender E F and Holdsworth P C W 1996 *Fluctuations and Order: The New Synthesis* ed M Millonas (Berlin: Springer) ch 16, p 259
[50] Plumer M L, Mailhot A and Caillé A 1994 *Phys. Rev.* B **49** 15 133
[51] Plumer M L and Mailhot A 1997 *J. Phys.: Condens. Matter* **9** L165
[52] Schotte U 1998 private communication