Quantum fluctuations in the incommensurate phase of $CsCuCl_3$ in a transverse magnetic field

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In zero magnetic field, the stacked, triangular antiferromagnet $CsCuCl_3$ has a helical structure incommensurate (IC) in the chain direction (normal to the planes). A magnetic field applied transverse to the chains distorts the helix, but the IC structure persists up to at least 0.43 times the saturation field. The IC wave number q (from neutron-diffraction experiments) decreases with increasing field, but then it has an unexpected plateau. Classical theory explains the behavior at small fields, including the temperature dependence, but it fails to explain the plateau, which we ascribe to quantum fluctuations. We find that linear spin-wave (LSW) theory also fails to explain the plateau; in fact, LSW theory fails more severely than classical theory in describing the IC phase. We introduce a phenomenological treatment of quantum fluctuations. After verifying that it describes well some known results, we apply the phenomenological theory to the IC phase of $CsCuCl_3$, finding that it yields a plateau at approximately the observed value of q and the observed fields; in addition, it predicts a transition to the commensurate phase so far not observed. Results depend sensitively on a weak anisotropy: A deviation of less than 1% from isotropy in the intrachain ferromagnetic exchange changes the phase diagram completely at fields above about half the saturation value. [S0163-1829(98)08409-4]

I. INTRODUCTION

Compounds of the ABX_3 family (A=Rb, Cs; B=Mn, Fe, Co, Ni, Cu, V; X=Cl, Br, I) figure prominently in the study of phase transitions in low-dimensional systems. Much of the interest in the magnetic-field behavior of compounds like $CsCuCl_3$ arises because they are physical realizations of models related to the triangular antiferromagnet (TAFM). Unlike the ground state of the square-lattice AFM, that of the TAFM is both continuously and discretely (twofold) degenerate, even in the presence of a magnetic field **H** (with magnitude H less than the saturation field H_S); the continuous degeneracy is nontrivial because it is not due to a symmetry of the Hamiltonian. In a field, thermal fluctuations[1–4] in classical TAFM models, and also quantum fluctuations,[5,6] break the continuous degeneracy in the same way, both selecting, for example, the colinear structure at $H \approx H_S/3$.

The magnetic properties of $CsCuCl_3$ (with a Néel temperature[7] $T_N = 10.7$ K) arise from the Cu^{2+} ions; to a good approximation, these form a triangular lattice of parallel chains, the other ions serving to define the structure. The major interactions, all nearest neighbor, are a ferromagnetic exchange interaction in the chain (c) direction, a weaker, frustrated, antiferromagnetic exchange interaction within the a-b planes, and a Dzyaloshinskii-Moriya[8] (DM) interaction also in the c direction; both exchange interactions are nearly isotropic. Recent studies[9,10] of the structure and of the phase transition giving rise to the DM term cite earlier literature on these topics. In the simplified structural model, the classical, zero-temperature, zero-field structure is a three-sublattice, $\pm 120°$ TAFM structure in each a-b plane; the spins lie in the planes and rotate from plane to plane, forming an incommensurate (IC) helical structure[11] with a pitch of $\approx 5.1°$.

Fluctuation effects are likely large in $CsCuCl_3$ for several reasons: The Cu spin is small ($S = 1/2$), the system is almost one dimensional (the intrachain interaction is significantly larger than the interchain interaction), the interchain interaction is frustrated, the exchange interactions are nearly isotropic, and the structure is incommensurate. Experiments in magnetic fields (difficult because $H_S = 30$ T) indeed find major effects due to fluctuations, both quantum and thermal. The quantum effects in $CsCuCl_3$ are very large; to our knowledge, they are exceeded in magnetic systems only by integer-spin vs half-integer-spin effects in nearly one-dimensional materials (including members of the ABX_3 family).

$CsCuCl_3$ in a longitudinal field ($\mathbf{H} \| c$) appears to be well understood at low temperatures T, but the transition at T_N has puzzling features.[12] The discontinuity[13–15] in the low-T magnetization at $H \approx 0.4\, H_S$ was shown by Shiba and Nikuni[16,17] to be a novel, fluctuation-induced phase transition from the umbrella structure (optimal at small H due to a small, easy-plane anisotropy[18] in the intrachain exchange) to a coplanar structure (optimal at larger H due to quantum fluctuations). Further experiments,[19–23] including neutron-diffraction[24] and specific-heat measurements[12] near T_N, confirmed their analysis.

Properties for a field transverse to the chains ($\mathbf{H} \perp c$) are not well established; such fields deform the helix, increasing its period. Classical theory[25,26] predicts that the IC wave number q decreases quadratically as H increases and that the curvature increases with T, both in agreement with experiment.[21,24] It predicts also a transition to a commensurate (C) phase (a stacked TAFM phase) at $H \approx 0.5\, H_S$. Experimentally, $CsCuCl_3$ is IC (Ref. 21) for $H \lesssim 0.43\, H_S$; its structure is unknown at larger H. Classical theory, which is sufficient for many materials, fails, however, to describe experimental results at fields near $H_S/3$ where structure is ob-

served in the magnetization[15] m, in the ^{133}Cs NMR shift,[23] in q (Ref. 21 finds a plateau), and in electron spin resonance (ESR) measurements.[22] The structure in m (only loosely a plateau) appears to result from quantum fluctuations; as for the TAFM,[5,6] linear spin-wave (LSW) theory finds[25] that the C-state magnetization has a plateau near $H_S/3$. Quantum fluctuations are surely responsible also for the plateau in q, but this remains to be demonstrated. Recent specific-heat, magnetization, and neutron-diffraction measurements[27] near T_N suggest major effects due to thermal fluctuations as well: T_N increases with field [as in the TAFM (Refs. 1, 3, and 28)] and a new phase appears.

This article develops the theory of the IC phase of $CsCuCl_3$ in a transverse field, with emphasis on the field dependence of q. We show that the natural, standard methods used to study such systems, namely classical theory and LSW theory (which adds the leading quantum correction), both fail to explain observed properties of $CsCuCl_3$; we know of few other systems for which both fail so badly. We introduce a phenomenological method to treat quantum fluctuations, establish its validity, show that it explains the plateau, and find that it predicts an IC→C transition.

Section II A describes the Hamiltonian. Section II B describes the classical (mean-field) theory, whose results are needed also for both the LSW and the phenomenological analyses. We include explicitly a small ($<1\%$) anisotropy in the intrachain exchange and we extend Ref. 25 also by examining out-of-plane solutions. We find that results are unexpectedly highly sensitive to the anisotropy, and that out-of-plane solutions can be optimal at intermediate fields (as found also by Jensen[29]). Section II C, which appears to be a technical advance (it describes LSW theory for a highly nonsinusoidal IC phase), finds that LSW theory not only fails to explain the plateau, it even worsens agreement with experiment. Going beyond LSW theory, to higher order in $1/S$, is out of the question. Section III introduces a phenomenological treatment of quantum fluctuations and shows that it describes some known properties of the TAFM and of the C states of $CsCuCl_3$. The same approach applied to the IC state yields a plateau in q (at approximately the observed value, over approximately the observed fields) followed by an IC →C transition (at a field well below the saturation field, likely below $0.5\,H_S$). See the note added at the end of the paper.

The failure of both classical theory and LSW theory (to describe the IC phase), and more generally the large fluctuation effects (both quantum and thermal), promise that $CsCuCl_3$ will reward further experimental and theoretical investigation.

II. HAMILTONIAN AND ANALYSIS

A. Hamiltonian

The Hamiltonian corresponding to the simplified structure is

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

where $\mathbf{S}_{in}$ is the spin operator at the ith site in the nth a-b plane, $\hat{\mathbf{z}}$ and $\hat{\mathbf{x}}$ are unit vectors in the c and a directions, and the k sum is over the six, in-plane, nearest neighbors of the site in. The first term ($\propto J_0$) is the isotropic, ferromagnetic exchange interaction between spins in nearest-neighbor planes, the second is an anisotropic correction (of easy-plane type) to the first, the third ($\propto D$) is the interplane DM interaction, the fourth ($\propto J_1$) is the isotropic, antiferromagnetic exchange interaction between nearest-neighbor spins in the a-b planes, and the fifth is the Zeeman energy in a transverse field $\mathbf{H}$. The coefficients have been estimated previously;[7,30,31,18,32] we use $J_0=28$ K, $\eta J_0=0.24$ K, $J_1=4.9$ K, and $D=5$ K. We omit the dipole-dipole interaction (which can induce an IC state modulated in the planes[33]), the anisotropy of the interchain interaction, and several small, related effects (namely, the displacement of the Cu ions from the c axis, the component of the DM vector perpendicular to the c axis, and the z component of the magnetizations[24,32]). The saturation field H_S, above which each spin is aligned with the field, is 30 T; for the isotropic model ($\eta=0$), $H_S=18J_1S/(g\mu_B)$.

At the classical level, the intrachain exchange term (J_0) favors states with spins parallel in adjacent a-b planes while the smaller DM term favors states with spins in the planes and rotating by $\pi/2$ per plane. The small anisotropy term[18] (responsible for the phase transition in longitudinal field[16,17]) also favors spins in the planes. The result at zero field is a helical structure: The spins lie in the planes, and the wave number is $q_0\hat{\mathbf{z}}$ with $q_0=\arctan[D/(2J_0)]\approx 2\pi/71$. At $H=0$, the continuous degeneracy of the classical TAFM ground state corresponds to a mere shift in the origin of the coordinate system; it will be important later that this degeneracy remains in a field (as long as $H<H_S$). A longitudinal field ($\mathbf{H}\|c$) maintains the axial symmetry of zero field; a layer-dependent rotation[16,17] in spin space eliminates the DM term, producing an easy-plane anisotropy. In contrast, a transverse field ($\mathbf{H}\perp c$) breaks the symmetry, and the DM term comes into full play.

B. Classical analysis

In the classical approximation, the spin operators $\mathbf{S}_{in}$ become classical vectors of length $S=1/2$, and the Hamiltonian of Eq. (1) becomes the energy function $E_{cl}(\{\mathbf{S}_{in}\})$. As found[21,24] at small H, the three-sublattice structure (indexed by $j=1,2,3$) is assumed for all $H<H_S$. The classical energy of the N spins is then

$$E_{cl}(\{\mathbf{S}_{jl}\})=\frac{N}{3L}\sum_{j=1}^{3}\sum_{l=1}^{L}\big[-2J_0\mathbf{S}_{jl}\cdot\mathbf{S}_{j,l+1}+2\eta J_0 S_{jl}^{(z)}S_{j,l+1}^{(z)} - D\hat{\mathbf{z}}\cdot(\mathbf{S}_{jl}\times\mathbf{S}_{j,l+1})+6J_1\mathbf{S}_{jl}\cdot\mathbf{S}_{j+1,l} - g\mu_B H\hat{\mathbf{x}}\cdot\mathbf{S}_{jl}\big], \tag{2}$$

where $l=1,\ldots,L$ is the layer index. The corresponding Euler-Lagrange equations are

$$-2J_0(\mathbf{S}_{j,l-1}+\mathbf{S}_{j,l+1})+2\eta J_0(S^{(z)}_{j,l-1}+S^{(z)}_{j,l+1})\hat{\mathbf{z}}$$
$$-D(\mathbf{S}_{j,l+1}-\mathbf{S}_{j,l-1})\times\hat{\mathbf{z}}+6J_1(\mathbf{S}_{j-1,l}+\mathbf{S}_{j+1,l})$$
$$-18J_1Sh\hat{\mathbf{x}}-2\lambda_{jl}\mathbf{S}_{jl}=0, \tag{3a}$$

where the λ_{jl} are Lagrange multipliers for the constraints

$$\mathbf{S}_{jl}\cdot\mathbf{S}_{jl}-S^2=0. \tag{3b}$$

These equations have many solutions. In the simplest, each layer has the same structure: The aligned state, with $\mathbf{S}_{jl}=S\hat{\mathbf{x}}$, is stable for $H\geqslant H_S$, and commensurate (C) states (stacked TAFM states, with three sublattices) exist for $H<H_S$. These states and their energies are easily obtained; the C states are continuously degenerate at the classical level, as for the TAFM. In contrast, the study of incommensurate (IC) states requires, in general, numerical solution of Eq. (3); the Appendix provides details.

The simplest IC state occurs at $H=0$: The spins lie in the planes ($\mathbf{S}_{jl}\perp c$), forming the 120° structure; the spins on the three sublattices rotate uniformly in the z direction. Many other IC states exist, particularly at intermediate H. Because the classical results are needed for the LSW analysis, which is possible only in wave number space, we retain the discrete model for the layer (l) dependence (avoiding the continuum approximation[25] which is nonetheless valid). We use periodic boundary conditions $\mathbf{S}_{j,l+L}=\mathbf{S}_{jl}$, with the IC-state period L limited to values $\lesssim 2000$. In principle, the periodic discrete model may miss some features of IC states, giving perhaps pinned structures and spurious energy gaps. We find, however, that the structures are not pinned (there are many degenerate solutions with the same period), and that the spin-wave energies are gapless at $\mathbf{k}=0$. This degeneracy is not the trivial one corresponding to a translation along the c axis, which in the discrete model corresponds to incrementing the layer index l.

For the IC states, it is natural to assume[25] that the spins lie in the planes at all fields, because then each term in the classical energy is either optimized (DM and ηJ_0 terms) or neutral [J_0 term and the sum of the interchain (J_1) and field terms]; that is, one assumes that the DM term confines the spins to the planes at all H (as it does at small H) and ignores the anisotropy term. These in-plane solutions were studied previously,[25] in the continuum approximation; in the simplest, either two or all three of the sublattice phases wind through 2π in one period, but there exist many other solutions which are composites of elementary solutions.[25] The solutions are sinusoidal at small H and solitonic at large. In an effort to understand some LSW results (Sec. II C), we investigated also out-of-plane IC solutions; surprisingly, these exist and can even be optimal at intermediate H, as found also in a study[29] of the mean-field equations at $T>0$. The Appendix argues that the out-of-plane solutions result from the frustration, and indeed we verified that in-plane solutions are optimal for all H in a model[34] with a ferromagnetic interchain interaction. Of course the anisotropy term favors in-plane solutions, as do quantum fluctuations (Sec. III), and out-of-plane solutions appear to have no consequences for $CsCuCl_3$, at least at $T=0$.

The main questions asked of the classical theory are (1) does a C state intervene between the IC state (known to be stable for $H=0$) and the aligned state (known to be stable for $H>H_S$), and (2) do the spins lie in the a-b planes at all H in the IC state? The answers depend crucially on the size of the anisotropy parameter η, a deviation of less than 1% from isotropy changing results dramatically.

For isotropic exchange ($\eta=0$), the structure is incommensurate up to H_S. For $h\lesssim 0.38$, there are many solutions (as before[25]), but they are well separated in energy; the optimal (in-plane) solution evolves continuously from the zero-field solution.[25] For $0.38\lesssim h<1$, out-of-plane solutions are optimal; the IC phase is very complicated, however (see the Appendix), and is best described as glassy in much of the range.

The easy-plane anisotropy[18] $\eta=8.6\times10^{-3}$ favors of course in-plane solutions; it changes the phase diagram substantially, opening a large window for the C phase and destroying the glassy phase, but it is too weak to destabilize out-of-plane solutions entirely. In more detail, the same in-plane IC solution is optimal for $0\lesssim h\lesssim 0.41$ (it extends to larger h because of the anisotropy), a simple, out-of-plane, IC solution is optimal for $0.42\lesssim h\lesssim 0.50$ (but other solutions are close in energy), and the C state is optimal at larger h; all transitions are second order. Likely an η value only slightly larger would destroy out-of-plane solutions at all h, giving a second-order IC→C transition[25] at $h\approx 0.47$.

C. Linear spin-wave analysis

To our knowledge, the following LSW analysis of the IC phase of $CsCuCl_3$ is the first study of quantum fluctuations in highly nonsinusoidal IC states. Of course spin waves in IC rare earths[35,36] have been studied for many years, and there is a large literature (Refs. 37–40 and references therein) on quantum effects in more general models of helimagnets. Much of the latter literature deals with the role of quantum fluctuations in breaking a classical degeneracy (to our knowledge, this role was first demonstrated by Shender[41]); in the axial next-nearest-neighbor Heisenberg model, for example, a degeneracy with respect to the IC wave number q exists in regions of parameter space, and quantum fluctuations (first examined in this context in Ref. 37) act to select q. Quantum fluctuations play a very different role in $CsCuCl_3$. The IC states of $CsCuCl_3$ are not classically degenerate in q and therefore no quantum selection among IC states occurs. In $CsCuCl_3$, quantum selection occurs instead for the C states, as in the TAFM. Part of the difficulty in treating $CsCuCl_3$ is then that LSW theory, which finds the leading correction ($\propto S^1$) to the classical energy ($\propto S^2$), does not determine the IC-state and C-state energies to the same accuracy. We argue below that the major effect of quantum fluctuations in the IC phase of $CsCuCl_3$ is to reconfigure the spins; this effect lies outside the scope of LSW theory.

We limit the analysis to in-plane spins and isotropic exchange ($\eta=0$), and so the field is limited to $h\lesssim 0.38$. LSW theory is so unsatisfactory that a more general quantitative analysis is unwarranted; qualitatively, $\eta>0$ favors in-plane spins and so results for $\eta=8.6\times10^{-3}$ would not differ substantially.

A Holstein-Primakoff transformation[42] to boson operators (using a local coordinate system[25]) and an expansion about the classical solutions give the Hamiltonian as

$$\mathcal{H}=E_{\rm cl}+\mathcal{H}_1+\mathcal{H}_2+{\rm O}(\sqrt{S}); \tag{4}$$

the classical spins are not colinear, and so the expansion parameter is $1/\sqrt{S}$ (rather than $1/S$), as in previous studies.[36,38,40,25,43] The classical angles ϕ_{jl} between the spins and the field are determined by minimizing the classical energy $E_{\rm cl}$ ($\propto S^2$):

$$E_{\rm cl}=S^2\frac{N}{3L}\sum_{j=1}^{3}\sum_{l=1}^{L}[-2J_0\cos(\phi_{j,l+1}-\phi_{jl})-D\sin(\phi_{j,l+1}-\phi_{jl})+6J_1\cos(\phi_{j+1,l}-\phi_{jl})-18J_1h\cos\phi_{jl}]. \tag{5}$$

The term $\mathcal{H}_1$ ($\propto S^{3/2}$) is linear in the boson operators; it vanishes for the classical angles ϕ_{jl} and so is omitted. The LSW Hamiltonian $\mathcal{H}_2$ ($\propto S$) has the standard form (quadratic in the boson operators b and $b^\dagger$)

$$\mathcal{H}_2=-\frac{S}{2}\frac{N}{3L}\sum_{jl}C_{jl}+\frac{S}{2}\sum_{\mathbf{k}}\sum_{jl}\sum_{j'l'}\{\mathcal{A}_{jl,j'l'}(\mathbf{k})\times[b_{jl}^\dagger(\mathbf{k})b_{j'l'}(\mathbf{k})+b_{jl}(-\mathbf{k})b_{j'l'}^\dagger(-\mathbf{k})]+\mathcal{B}_{jl,j'l'}(\mathbf{k})\times[b_{jl}^\dagger(\mathbf{k})b_{j'l'}^\dagger(-\mathbf{k})+b_{jl}(-\mathbf{k})b_{j'l'}(\mathbf{k})]\}. \tag{6}$$

The $\mathbf{k}$ sum in the second term runs over the $N/(3L)$ points in the first Brillouin zone; the $3L\times 3L$ Hermitian matrices $\mathcal{A}$, $\mathcal{B}$, and $\mathcal{C}$, the last diagonal ($\mathcal{C}_{jl,j'l'}=C_{jl}\delta_{jj'}\delta_{ll'}$) and independent of $\mathbf{k}$, are

$$\begin{aligned}[\mathcal{A}(\mathbf{k})+\mathcal{B}(\mathbf{k})-\mathcal{C}]_{jl,j'l'}&=-2J_0\cos(\phi_{jl'}-\phi_{jl})\delta_{j'j}(e^{ik_z}\delta_{l',l+1}+e^{-ik_z}\delta_{l',l-1})\\&\quad-D\sin(\phi_{jl'}-\phi_{jl})\delta_{j'j}(e^{ik_z}\delta_{l',l+1}-e^{-ik_z}\delta_{l',l-1})\\&\quad+6J_1\cos(\phi_{j'l}-\phi_{jl})\\&\quad\times\delta_{l'l}(\nu_{\mathbf{k}}\delta_{j',j+1}+\nu_{\mathbf{k}}^*\delta_{j',j-1}),\end{aligned} \tag{7}$$

$$\begin{aligned}[\mathcal{A}(\mathbf{k})-\mathcal{B}(\mathbf{k})-\mathcal{C}]_{jl,j'l'}&=-2J_0\delta_{j'j}(e^{ik_z}\delta_{l',l+1}+e^{-ik_z}\delta_{l',l-1})+6J_1\delta_{l'l}(\nu_{\mathbf{k}}\delta_{j',j+1}+\nu_{\mathbf{k}}^*\delta_{j',j-1}),\end{aligned} \tag{8}$$

$$\begin{aligned}C_{jl}&=2J_0[\cos(\phi_{j,l+1}-\phi_{jl})+\cos(\phi_{jl}-\phi_{j,l-1})]\\&\quad+D[\sin(\phi_{j,l+1}-\phi_{jl})+\sin(\phi_{jl}-\phi_{j,l-1})]\\&\quad-6J_1[\cos(\phi_{j+1,l}-\phi_{jl})+\cos(\phi_{j-1,l}-\phi_{jl})]\\&\quad+18J_1h\cos\phi_{jl},\end{aligned} \tag{9}$$

where $\nu_{\mathbf{k}}$ is the in-plane structure factor,

$$\nu_{\mathbf{k}}=\tfrac{1}{3}\{\exp(ik_x)+\exp[\tfrac{1}{2}i(-k_x+\sqrt{3}k_y)]+\exp[\tfrac{1}{2}i(-k_x-\sqrt{3}k_y)]\}. \tag{10}$$

Note that $\mathcal{H}_2$ is determined by the classical angles ϕ_{jl}. A standard transformation[42,44] to creation and annihilation operators $\gamma_{jl}^\dagger$ and γ_{jl} for the spin-wave excitations gives

$$\mathcal{H}_2=E_2+S\sum_{\mathbf{k}}\sum_{jl}\epsilon_{jl}(\mathbf{k})\gamma_{jl}^\dagger(\mathbf{k})\gamma_{jl}(\mathbf{k}), \tag{11}$$

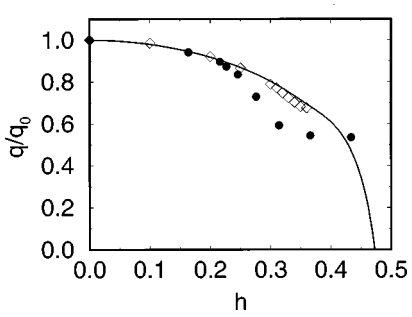

FIG. 1. Dependence of the reduced wave number q/q_0 on the reduced field h. The solid circles are the experimental results of Ref. 21. Both the classical result (solid line, from Ref. 25) and the linear spin-wave result (diamonds) were found for in-plane spins and isotropic exchange ($\eta=0$); the latter were obtained only for $h\leqslant 0.38$, as described in the text.

where the ground-state value $E_2=\langle 0|\mathcal{H}_2|0\rangle$ is

$$E_2=-\frac{S}{2}\frac{N}{3L}\sum_{jl}C_{jl}+\frac{S}{2}\sum_{\mathbf{k}}\sum_{jl}\epsilon_{jl}(\mathbf{k}). \tag{12}$$

The excitation (spin-wave) energies ϵ ($\geqslant 0$) are found from

$$\det[(\mathcal{A}-\mathcal{B})(\mathcal{A}+\mathcal{B})-\epsilon^2\hat{1}]=0; \tag{13}$$

this eigenvalue problem for $\epsilon_{jl}^2(\mathbf{k})$ is Hermitian, but the result for $\epsilon_{jl}(\mathbf{k})$ can be imaginary (as when in-plane solutions become unstable at intermediate fields).

The LSW analysis of quantum fluctuations in the C state is straightforward;[25] of course the antisymmetric DM term does not appear, and so quantum selection occurs just as in the TAFM.[6] For the IC state (the 111 state of Ref. 25), the total energy $E_{\rm cl}+E_2$ was found as a function of the period L and minimized with respect to L to give the optimal wave number $q=2\pi/L$; the Brillouin-zone integration in Eq. (12) was performed using 500–1000 points. In the relevant field region, the optimal L ranged from ≈ 70 a-b plane spacings to ≈ 150, large enough to justify use of the discrete model and small enough that the diagonalization was practicable.

Figure 1 shows that LSW theory (like classical theory[25]) fails to describe the experiment results[21] for q, although the LSW results may flatten slightly with increasing field. Far more seriously, LSW theory predicts an unobserved transition to the C phase at $h\approx 0.32$, and so its predictions at larger h are irrelevant. That is, LSW theory provides a worse description than does classical theory, by predicting a C phase to exist for fields where experiment finds, and classical theory predicts, an IC phase.

The failure of LSW theory to describe the IC phase of $CsCuCl_3$ at intermediate fields deserves further comment. There are two points here. First, LSW theory predicts the premature IC→C transition (at $h\approx 0.32$) because it finds the C-state energy more accurately than it does the IC-state energy. The C-state energy is found by choosing the classical configuration (for example, the colinear state at $h=1/3$) which minimizes the total energy $E_{\rm cl}+E_2$, but such a choice is not possible for the IC state. Second, LSW theory fails for the more profound reason that the classical IC state is a poor approximation to the quantum IC state: LSW theory does not take into account the breaking of the continuous degeneracy in determining the spin structure of the IC phase. Conven-

tional IC phases have domain walls where the order parameter passes rapidly through commensurately forbidden values, from one commensurately allowed value to another. The classical, in-plane IC structure of $CsCuCl_3$ is very different;[25] it can be described instead as a sequence of degenerate C states (loosely, a ''spatially varying commensurate state''); domain walls are well defined only very close to the IC-C transition.[25] But quantum fluctuations destroy the continuous degeneracy and so the quantum IC state at even small fields will have the conventional IC structure described above. The quantum structure is very different from the classical one, and thus is difficult to obtain by the perturbative approach of the $1/S$ expansion.

We believe that extension of the analysis to higher order (including the spin rearrangement due to quantum fluctuations, as in Refs. 38, 40, and 43) would be futile; for example, it would still be difficult to find the two energies to the same accuracy. Further, the extension is computationally out of reach, entirely without appeal, and even not possible if $\eta=0$ (the classical ground state cannot be found for $h\gtrsim 0.38$).

A very different approach is required.

III. PHENOMENOLOGICAL TREATMENT OF QUANTUM FLUCTUATIONS

The most important effect of quantum fluctuations in this problem is the breaking of the TAFM classical degeneracy. The TAFM part of the Hamiltonian of Eq. (1) contributes to the energy at both the classical level (previous section) and the quantum level. The following describes an innovative method to treat the quantum contribution, establishes its validity, and applies it to $CsCuCl_3$ in transverse field.

The innovation is to represent the quantum contribution phenomenologically by the term

$$E_{\rm fluct}=-\frac{J_2}{S^2}\sum_{\langle ij\rangle}\sum_{n}(\mathbf{S}_{in}\cdot\mathbf{S}_{jn})^2, \tag{14a}$$

where the first sum is over nearest-neighbor pairs; for in-plane spins, $E_{\rm fluct}$ simplifies to

$$E_{\rm fluct}=-J_2S^2\frac{N}{L}\sum_{j=1}^{3}\sum_{l=1}^{L}\cos^2(\phi_{j+1,l}-\phi_{jl}). \tag{14b}$$

The total energy, a function of the classical vectors $\mathbf{S}_{in}$, is $E_{\rm total}=E_{\rm cl}+E_{\rm fluct}$. The classical energy $E_{\rm cl}$ is proportional to S^2; the fluctuation energy $E_{\rm fluct}$ contains, in principle, all higher-order terms in the $1/S$ expansion. The coefficient J_2 is positive (quantum fluctuations reduce the energy of, say, the colinear state near $h=1/3$ in the TAFM) and is essentially a free parameter, although it can be estimated as described below; it vanishes as $1/S$ as $S\to\infty$, but contains in principle higher-order terms as well.

Note that we do not add a biquadratic coupling term to the Hamiltonian; such a term cannot contribute for a $S=1/2$ system like $CsCuCl_3$, and its appearance in the expression for the total energy is therefore at the phenomenological level. Of course such terms appear in the Hamiltonians of systems with $S>1/2$; they have been studied previously (as in the classical theory[45] of the plateau[46] in the magnetization of the $S=7/2$ system C_6Eu), and Ref. 6 pointed out that such a term may have the same consequences as quantum fluctuations. More to the point, a biquadratic coupling term has appeared explicitly in analytical treatments of quantum (and thermal) fluctuations.[41,47–51]

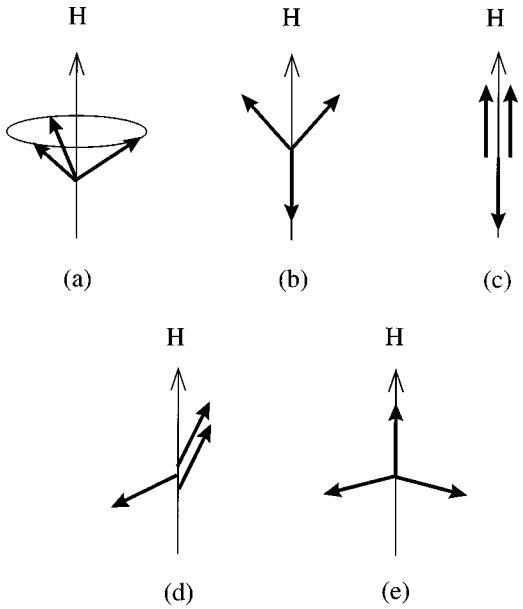

FIG. 2. The five states, the umbrella state (a) and the four coplanar states (b)–(e), used in the examination of the phenomenological energy $E_{\rm fluct}$.

The following tests the phenomenological theory by comparing its predictions with those of the $1/S$ expansion, for the TAFM. We minimized the total phenomenological energy

$$E_{\rm total}=N\sum_{j=1}^{3}[2J_1\mathbf{S}_j\cdot\mathbf{S}_{j+1}-g\mu_{\rm B}\mathbf{H}\cdot\mathbf{S}_j/3-J_2(\mathbf{S}_j\cdot\mathbf{S}_{j+1})^2/S^2] \tag{15}$$

to determine the spin configurations and the energies for the five high-symmetry TAFM states of Fig. 2; recall that the classical TAFM states are continuously degenerate, also with respect to the direction of **H**. We omit the expressions (easily obtained) for the energies, quoting only the following results.

(a) The umbrella state: The angle ϕ between the spins and the field is found from

$$18J_1S\cos\phi-9J_2S\cos\phi(3\cos^2\phi-1)=g\mu_{\rm B}H. \tag{16}$$

This state exists for all $H<H_{\rm S}$; it is optimal at no field >0.

(b) The coplanar state $\phi_1=\pi$, $\phi_3=-\phi_2$ with $\sin\phi_2\neq 0$: The angle ϕ_2 is found from

$$6J_1S(2\cos\phi_2-1)-6J_2S\cos\phi_2(1+2\cos2\phi_2)=g\mu_{\rm B}H. \tag{17}$$

This state exists, and is optimal, for $0<H<H_1=6(J_1-3J_2)S/g\mu_{\rm B}$.

(c) The colinear state $\phi_1=\pi$, $\phi_2=\phi_3=0$. This state is optimal at intermediate fields, $H_1<H<H_2=6(J_1+J_2)S/g\mu_{\rm B}$; the magnetization is one-third the saturation value.

(d) The coplanar state $\phi_3=\phi_2$ with $\sin\phi_2\neq 0$: The angles ϕ_1 and ϕ_2 are determined by

$$12\,J_1 S\sin(\phi_1-\phi_2)-6\,J_2 S\sin(2\phi_1-2\phi_2)=g\mu_B H\sin\phi_1, \tag{18a}$$

$$\sin\phi_1+2\sin\phi_2=0. \tag{18b}$$

This state exists, and is optimal, for $H_2<H<H_S=18(J_1-J_2)S/g\mu_B$.

(e) The coplanar state $\phi_1=0$, $\phi_3=-\phi_2$ with $\sin\phi_2\neq 0$: The angle ϕ_2 is found from

$$6\,J_1 S(2\cos\phi_2+1)-6\,J_2 S\cos\phi_2(1+2\cos 2\phi_2)=g\mu_B H. \tag{19}$$

This state exists for all $H<H_S$; it is optimal at no field >0.

The phenomenological theory then predicts the following phase sequence as H is increased: state (b) $\rightarrow$ state (c) $\rightarrow$ state (d) $\rightarrow$ aligned state ($\phi_j=0$); it predicts also a plateau in the magnetization for $H_1<H<H_2$. These results agree in all respects with those of the microscopic $(1/S)$ theory[6] for quantum fluctuations in the TAFM. There are some differences with respect to the transition fields H_1, H_2, and H_S: First, quantum fluctuations do not renormalize[6] H_S, which remains $18J_1S/(g\mu_B)$ in the isotropic model. Second, the phenomenological theory gives $H_1/H_S\approx(1-2J_2/J_1)/3$ and $H_2/H_S\approx(1+2J_2/J_1)/3$, whereas LSW theory[6] gives $H_1/H_S\approx(1-0.084/S)/3$ and $H_2/H_S\approx(1+0.215/S)/3$; the first are symmetrical about 1/3, the second not. Comparison of the expressions for H_1 and H_2 gives the rough estimate $2SJ_2/J_1\approx 0.1$–0.2, but comparison of the energies may give a different value; in any case, these estimates apply to the TAFM only, and the value appropriate to other models may be significantly different.

Having shown that the phenomenological theory provides a good qualitative description of quantum fluctuations in the TAFM, we next test it quantitatively for the energies of the five C states of $CsCuCl_3$ derived from the TAFM states of Fig. 2. The phenomenological energies $E_{\rm fluct}$ are the same as for the TAFM and are independent of J_0. The LSW energies E_2 ($\propto S$), which depend on J_0, were found[17] by standard methods [see Eq. (12)] using the parameter values $J_0=28$ K, $\eta=0$, $J_1=4.9$ K and $S=1/2$; a separate comparison should really be made for $\eta=8.6\times10^{-3}$ but this seems unwarranted. To make a fair comparison (E_2 is only the leading quantum correction), we found $E_{\rm fluct}$ to only first order in J_2; this means, for example, that $\cos\phi=h=g\mu_B H/18J_1S$ for the umbrella state. Figure 3 plots the two energies for states (a)–(d), relative to state (e) (which exists over the entire range $0\leq H<H_S$ and so makes a convenient reference state). Good agreement is obtained for $J_2=0.2$ K ($2SJ_2/J_1\approx 0.04$) and so we expect that the phenomenological term $E_{\rm fluct}$ captures well enough the breaking of the classical degeneracy by quantum fluctuations.[52] Our previous investigation[25] of the C-state spin configurations gave the results $H_1/H_S\approx 0.31$ and $H_2/H_S\approx 0.37$; these correspond to $2SJ_2/J_1=0.03_5$ and 0.05_5, respectively, in reasonable agreement with the above value (0.04). Because E_2 is only the first-order correction, a moderate adjustment of J_2 is acceptable; in fact, an effective J_2 smaller than 0.2 K is suggested by the next correction[53] at fields just below H_S.

The phenomenological approach was then applied to the IC phase of $CsCuCl_3$ in a transverse field. The Euler-Lagrange equations for the total energy $E_{\rm cl}+E_{\rm fluct}$ are

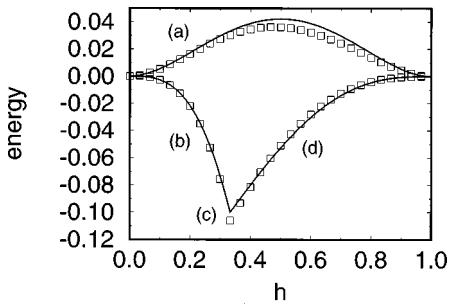

FIG. 3. Comparison of the linear spin-wave result E_2 (squares) and the phenomenological energy $E_{\rm fluct}$ (lines, with $J_2=0.2$ K, to first order in J_2) for the five commensurate $CsCuCl_3$ states corresponding to the states of Fig. 2. The figure gives the energies E_a-E_e, etc., of states (a)–(d) relative to that of state (e); the energy of state (b) is plotted only for $h<1/3$, that of (c) only for $h=1/3$, and that of (d) only for $h>1/3$.

$$\begin{aligned}&-2\,J_0(\mathbf{S}_{j,l-1}+\mathbf{S}_{j,l+1})+2\eta J_0(S^{(z)}_{j,l-1}+S^{(z)}_{j,l+1})\hat{\mathbf{z}}-D(\mathbf{S}_{j,l+1}\\&\quad-\mathbf{S}_{j,l-1})\times\hat{\mathbf{z}}+6\,J_1(\mathbf{S}_{j-1,l}+\mathbf{S}_{j+1,l})-g\mu_B H\hat{\mathbf{x}}-2\lambda_{jl}\mathbf{S}_{jl}\\&\quad-6\,J_2 S^{-2}[(\mathbf{S}_{j-1,l}\cdot\mathbf{S}_{jl})\mathbf{S}_{j-1,l}+(\mathbf{S}_{j+1,l}\cdot\mathbf{S}_{jl})\mathbf{S}_{j+1,l}]=0,\end{aligned} \tag{20}$$

plus the constraints of Eq. (3b); for in-plane spins, these simplify to

$$\begin{aligned}&-2\,J_0 S[\sin(\phi_{jl}-\phi_{j,l-1})+\sin(\phi_{jl}-\phi_{j,l+1})]-DS[\cos(\phi_{jl}\\&\quad-\phi_{j,l+1})-\cos(\phi_{jl}-\phi_{j,l-1})]+6\,J_1 S[\sin(\phi_{jl}\\&\quad-\phi_{j-1,l})+\sin(\phi_{jl}-\phi_{j+1,l})]-g\mu_B H\sin\phi_{jl}\\&\quad-3\,J_2 S[\sin(2\phi_{jl}-2\phi_{j-1,l})+\sin(2\phi_{jl}-2\phi_{j+1,l})]=0.\end{aligned} \tag{21}$$

Scores of solutions of these equations were studied, as described in the Appendix. We use $J_2=0.13$ K, as discussed below, and define the reduced field as $h=g\mu_B H/(18J_1S)$; note that saturation occurs slightly below $h=1$. The phenomenological term has no effect at $H=0$ and only weak effects at small H, but major differences are found for $H\gtrsim H_S/3$.

For isotropic exchange ($\eta=0$), the ground state is incommensurate up to H_S. The in-plane solution[25] is optimal for $0\leq h\leq 0.38$; above this field, two out-of-plane solutions compete, their energies crossing several times. That is, quantum fluctuations destroy the glassy phase of Sec. II B.

More appropriate for $CsCuCl_3$ are results for the experimental anisotropy ($\eta=0.0086$). Including quantum fluctuations at the best level presently possible, by means of the phenomenological theory, we find the results of Fig. 4 for the IC wave number q. The agreement with experiment[21] is moderately good: The plateau occurs at about the observed value of q/q_0 and over about the correct field range; although measurements are available only to $H=13$ T or $h=0.43$, it appears, however, that the theoretical value drops prematurely. As before,[25] the IC$\rightarrow$C transition (here at $h\approx 0.44$) occurs because in-plane spins must pass through high-energy (commensurately forbidden) configurations as they wind; out-of-plane spins are disfavored by the aniso-

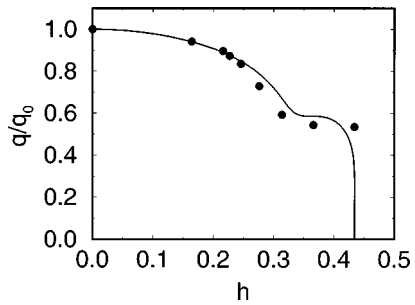

FIG. 4. Dependence of the reduced wave number q/q_0 on the reduced field h. The circles are the experimental results of Ref. 21; the structure is unknown beyond $h \approx 0.43$. The line gives the theoretical results based on a phenomenological treatment of quantum fluctuations ($J_2 = 0.13$ K), for weak easy-plane anisotropy ($\eta = 8.6 \times 10^{-3}$); the commensurate state is stable for $h > 0.44$.

tropic exchange and the quantum fluctuations, as well as by the DM term. The magnetization is moderately rounded near $h = 1/3$, as in experiment,[15] but the agreement is not quantitative. The value $J_2 = 0.13$ K was chosen because it gives a plateau in q, but neither the level of the plateau nor the field range over which it occurs is adjustable; no plateau is found for significantly smaller J_2, and q is nonmonotonic for significantly larger J_2 (0.17 K for example).[54] In more detail, the in-plane IC solution[25] is optimal for $h \leqslant 0.42$, and the C phase is optimal from $h \geqslant 0.44$ up to $H = H_S$; an out-of-plane IC solution is optimal over a small field range ($\Delta h < 0.013$) about $h = 0.43$, but we consider that it should not be taken seriously, given the sensitivity to the value of η.

Note added. Six months after submitting this manuscript, we were informed that neutron-diffraction experiments[55] (with both pulsed field and pulsed neutrons) have observed the IC→C transition predicted by the phenomenological theory; differences are that, experimentally, the wave number drops less precipitously, and the transition occurs at $H \approx 0.58\, H_S$, ''well below the saturation field'' (as in the Introduction), but larger than our $0.44\, H_S$ or ''likely below $0.5\, H_S$.'' Likely the phenomenological theory can be improved.

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APPENDIX

The following provides details regarding the solution of the Euler-Lagrange equations (3) and the extension (20).

For given period L and reduced field $h = H/H_S$, the $12L$ equations (for $\mathbf{S}_{jl}$ and λ_{jl}) were solved by linearization about trial solutions and solution of the resulting linear equations, repeated to convergence; the trial solutions were obtained from solutions at nearby fields, by interpolation in solutions at nearby periods, and by other means. Because the IC solutions are not pinned (and the equations have many nontrivially degenerate solutions), an additional condition such as $\hat{\mathbf{y}} \cdot \mathbf{S}_{11} = 0$ must be imposed; without it the linear equations are indeterminate. The extra condition results from the method used to solve the Euler-Lagrange equations; it is unnecessary when the energy is minimized directly. Because the period is discrete (L is an integer), the accuracy to which the energy and the wave number $q = 2\pi/L$ can be determined is limited, but better values are easily found by interpolation or by generating composite solutions (join two or more identical solutions to get a new solution and minimize its energy with respect to the new period).

In general, the number of out-of-plane IC solutions increases enormously for $h \gtrsim 0.4$, and the question arises whether one can be reasonably confident that the optimal (lowest-energy) solution has been found; guides are whether the solution is simple, whether it is optimal for a reasonably large range of field, and whether other solutions are well separated in energy. Too often we found the optimal solution to be neither simple nor obvious, and so it was necessary to generate and investigate scores of solutions. These were generated at various values of h and L as follows: Random values were assigned to the phase angles; approximate solutions were found by conjugate-gradient minimization of the energy; refined solutions were obtained from the Euler-Lagrange equations. A solution found in this way was optimized with respect to L, at fixed h. It was then followed as a function of h, the solution at one period L and reduced field h being used as a trial solution at the same L and a field $h \pm 0.01$; at each h, the energy of the solution was minimized with respect to L.

For isotropic intraplane exchange ($\eta = 0$), and without the term representing quantum fluctuations, the IC phase is glassy in most of the range $h > 0.38$; the optimal solution cannot be identified with any confidence. The solutions are far more numerous than at smaller fields; they differ only slightly in energy for the most part; the solution optimal at one field is often not optimal at nearby fields; solutions stable at one field are often unstable at nearby fields; the energies of solutions often cross as the field is changed; at a given field, the optimal solution is often complicated; and many new solutions are found with additional random starting configurations at different values of h and L.

The existence of out-of-plane IC solutions, and that they can be optimal, can be traced, it appears, to the classical degeneracy of the TAFM. Recall that the TAFM condition $\Sigma_{j=1}^{3} \mathbf{S}_j = 3\, S\mathbf{H}/H_S$ for $H \leqslant H_S$ gives only three conditions for the six variables needed to specify the spins on the three sublattices; some of this great freedom may carry over to the classical IC structure of $CsCuCl_3$. This interpretation suggests that spins should remain in the planes if the interchain interaction is not frustrated; we verified this for a particular model.[34] Because quantum fluctuations in the TAFM favor coplanar states (with the spins and the field in the same plane) over noncoplanar states (such as the umbrella state), the interpretation suggests also, as found by the phenomenological treatment of Sec. III, that quantum fluctuations (treated at the proper level—this means going beyond LSW theory for the IC phase of $CsCuCl_3$) should disfavor out-of-plane solutions. The anisotropy and quantum fluctuations combine to eliminate out-of-plane solutions almost entirely. Finally, we mention that the argument (Sec. II B) for in-plane solutions fails because the in-plane IC structure forces the spins to wind through high-energy configurations when $h > 1/3$; for out-of-plane solutions, some spins can instead oscillate about a commensurate value, giving a rippled commensurate state.

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