

Scaling analysis of the new multicritical behavior of CsMnBr_3 and CsNiCl_3

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Critical properties of the axial triangular antiferromagnet CsNiCl_3 and the planar triangular antiferromagnet CsMnBr_3 under an applied magnetic field are studied using scaling theory. Novel types of multicritical behavior observed in these compounds are associated with $n=2$ (CsMnBr_3) and $n=3$ (CsNiCl_3) chiral universality classes recently identified by the renormalization-group analysis. Various experimentally testable predictions are derived on the basis of a scaling ansatz combined with the numerical estimates of critical exponents. In particular, all phase boundaries emanating from the multicritical points are found to be scaled by the corresponding anisotropy-crossover exponent. It is also predicted that the criticality along the high-field critical line of CsNiCl_3 (paramagnetic to spin flop) is of $n=2$ chiral universality.

I. INTRODUCTION

There has been continuous interest in the magnetic properties of a large class of hexagonal insulators with the chemical formula ABX_3 , where B represents magnetic ions such as Ni, Cu, Co, Cr, and Fe. In this class of materials, the magnetic ions form linear chains along the c axis that are coupled antiferromagnetically forming triangular layers in the basal a - b plane. Although these compounds are magnetically quasi-one-dimensional, most of them are known to undergo magnetic phase transitions into a *three-dimensionally* ordered state at low temperatures driven by the weak interchain coupling. Thus, magnetic frustration, which arises because of the antiferromagnetic nature of the interchain coupling combined with the triangular-lattice structure, is expected to play a crucial role in their magnetic structure. In fact, the nature of the magnetic ordering of these ABX_3 compounds are often quite novel, markedly different from those of usual ferromagnets or unfrustrated antiferromagnets on bipartite lattices.

If the long-range dipolar interaction is sufficiently weak, which appears to be the case when the intrachain coupling is antiferromagnetic, the magnetic ordering of these compounds may roughly be classified into two categories depending on the type of their magnetic anisotropies. If the magnetic ion has an easy-axis-type anisotropy, the compound exhibits two successive transitions in zero applied field, say at $T=T_{N1}$ and T_{N2} ($T_{N1} > T_{N2}$). A typical example of an axial triangular antiferromagnet is CsNiCl_3 , which has Néel temperatures at $T_{N1} \approx 4.85$ K and $T_{N2} \approx 4.40$ K.¹⁻³ At $T=T_{N1}$, only the longitudinal (c axis) component orders and a linearly-polarized spin structure is realized for $T_{N2} < T < T_{N1}$. The transverse components order at $T=T_{N2}$ and the elliptically polarized structure is stabilized for $T < T_{N2}$.^{1,2} The magnetic field-temperature phase diagram of CsNiCl_3 was experimentally determined by Johnson, Rayne, and Friedberg

by means of the susceptibility measurements.³ Their results, obtained with the magnetic field applied along the easy c axis, is schematically reproduced in Fig. 1. The most striking feature revealed by their experiment is the existence of a novel type of multicritical point at a finite field, at which a line of first-order transitions (spin-flop line) and three distinct critical lines meet. In fact, it was shown by Plumer, Hood, and Caillé that such an unusual type of multicritical point is possible within the framework of the Landau theory.⁴ The nature of the spin ordering in each phase was also determined (see Fig. 1).

On the other hand, if the magnetic ion has an easy-plane-type anisotropy, the compound exhibits a single transition in zero applied field at $T=T_N$. A typical example of such planar triangular antiferromagnets is CsMnBr_3 , which has a Néel temperature at $T_N \approx 8.3$ K.⁵⁻⁷ The spin ordering at $T < T_N$ is the so-called 120°

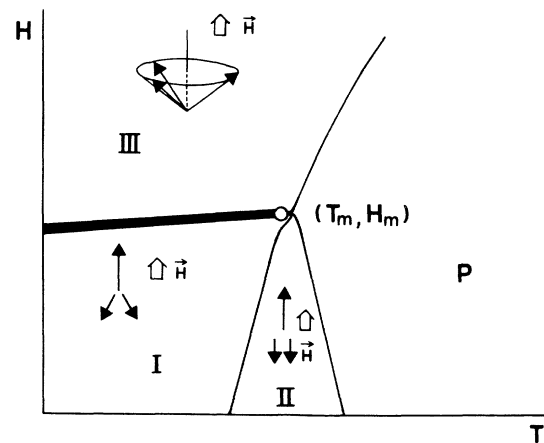


FIG. 1. Schematic magnetic phase diagram of CsNiCl_3 . Magnetic field is applied along an easy c axis. The I-III boundary line represents the first-order spin-flop transition.

spin structure in the basal plane. The magnetic field-temperature phase diagram of CsMnBr_3 was experimentally determined by Gaulin *et al.* by use of elastic neutron scattering.⁷ Their result, obtained with the magnetic field applied in the basal plane, is schematically shown in Fig. 2. The single phase transition observed in zero field was found to split into two successive transitions under finite fields. Thus, the Néel point in zero field has turned out to be a *tetracritical point*. Subsequently, a Landau-type free-energy analysis was applied to CsMnBr_3 by Plumer and Caillé, who successfully reproduced the experimentally-observed magnetic phase diagram including the tetracritical point at zero field and determined the type of spin ordering in each phase as shown in Fig. 2.⁸

Criticality of the zero-field transition of CsMnBr_3 has attracted considerable interest because of a recent theoretical prediction by Kawamura that the chiral degeneracy associated with the helical spin ordering might lead to a new universality class,⁹ distinct from the conventional $O(n)$ Heisenberg universality class. Subsequent experiments carried out on CsMnBr_3 ,^{5,6} and several other materials including VCl_2 ,¹⁰ VBr_2 ,¹¹ Ho ,¹² and Dy ,¹² seem to support this theoretical prediction. If this is really the case, tetracritical behavior of CsMnBr_3 should be described by a new type of “chiral” universality.

In the present paper, we perform a detailed scaling theory analysis of the critical behavior of CsMnBr_3 and CsNiCl_3 under applied magnetic fields, with major emphasis on elucidating the novel multicritical behavior observed experimentally in these compounds. We start with the Ginzburg-Landau Hamiltonian used by previous authors, but go beyond the mean-field level by taking into account the effect of critical fluctuations. The case of CsMnBr_3 is analyzed in Sec. II. The zero field tetracriticality found in this material is ascribed to the $n=2$ chiral universality. At sufficiently small fields, two critical lines emanating from the tetracritical point are predicted to be scaled by the corresponding anisotropy-crossover exponent. The case of CsNiCl_3 is analyzed in Sec. III.

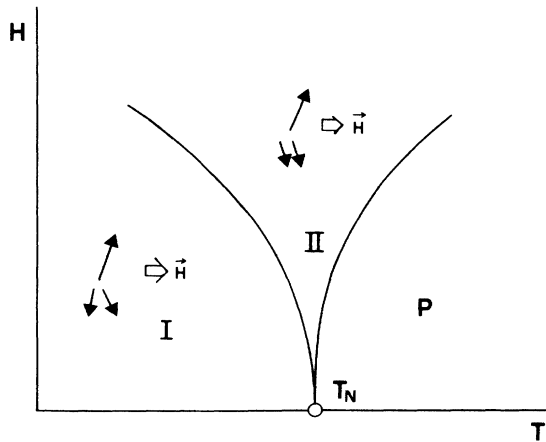


FIG. 2. Schematic magnetic phase diagram of CsMnBr_3 . Magnetic field is applied in an easy a - b plane.

Novel type of multicriticality observed at a finite field is ascribed to the $n=3$ chiral universality. In the vicinity of this multicritical point, all three critical lines are found to be scaled by a common anisotropy-crossover exponent associated with $n=3$ chiral class. We also predict that the criticality along the high-field critical line between the paramagnetic and the spin-flop phases should be of $n=2$ chiral universality. Based on the scaling *ansatz*, various experimentally testable predictions are derived for both materials. Finally in Sec. IV, we discuss the experimental implications of the obtained results.

II. SCALING ANALYSIS OF CsMnBr_3

In this section, we analyze the case of CsMnBr_3 . A magnetic field is assumed to be applied in the basal plane as $\mathbf{H}=(H,0)$, and only the basal-plane components will be considered. The appropriate Ginzburg-Landau-Wilson (GLW) Hamiltonian is given by^{8,9(b)}

$$\mathcal{H}=\mathcal{H}_0+\mathcal{H}_m, \quad (1)$$

$$\begin{aligned} \mathcal{H}_0 &= (\nabla \mathbf{a})^2 + (\nabla \mathbf{b})^2 + r_0(\mathbf{a}^2 + \mathbf{b}^2) \\ &+ u(\mathbf{a}^2 + \mathbf{b}^2)^2 + v[(\mathbf{a} \cdot \mathbf{b})^2 - \mathbf{a}^2 \mathbf{b}^2], \end{aligned} \quad (2)$$

$$\begin{aligned} \mathcal{H}_m &= c\mathbf{m}^2 + d\mathbf{m}^4 + e(\mathbf{a}^2 + \mathbf{b}^2)\mathbf{m}^2 \\ &+ f[(\mathbf{a} \cdot \mathbf{m})^2 + (\mathbf{b} \cdot \mathbf{m})^2] - \mathbf{H} \cdot \mathbf{m}, \end{aligned} \quad (3)$$

where \mathbf{m} is the uniform magnetization induced by an applied magnetic field, and the two vector fields $\mathbf{a}=(a_x, a_y)$ and $\mathbf{b}=(b_x, b_y)$ represent the cosine and sine modes associated with the wave vector $\mathbf{Q}=(4\pi/3, 0, \pi)$ via,

$$\mathbf{s}(\mathbf{r}) = \mathbf{m} + \mathbf{a} \cos(\mathbf{Q} \cdot \mathbf{r}) + \mathbf{b} \sin(\mathbf{Q} \cdot \mathbf{r}). \quad (4)$$

The first term in (1), \mathcal{H}_0 , describes CsMnBr_3 in zero field, while the second term, \mathcal{H}_m , contains the terms induced by the applied field.

Plumer and Caillé performed a Landau-type free-energy analysis for the Hamiltonian (1), which neglects the effects of fluctuations.⁸ With an appropriate choice of the parameters, $u > 0$, $v > 0$, $f > 0$, $e < 0$, etc., these authors successfully reproduced the experimentally observed magnetic phase diagram of CsMnBr_3 .⁷ According to this analysis, the spin ordering in zero field is a helically polarized state (120° spin structure), corresponding to $\mathbf{a} \perp \mathbf{b}$ with $|\mathbf{a}| = |\mathbf{b}|$. At finite fields, two successive transitions occur: the spin ordering in the intermediate phase (phase II in Fig. 2) is a linearly polarized state with $\mathbf{a} \perp \mathbf{H}$ and $\mathbf{b} = 0$ (or equivalently, $\mathbf{b} \perp \mathbf{H}$ and $\mathbf{a} = 0$), while that in the low-temperature phase (phase I) is an elliptically polarized state with $\mathbf{a} \perp \mathbf{b}$ and $|\mathbf{a}| \neq |\mathbf{b}|$.

We now further examine the nature of the phase transition beyond the Landau theory by taking into account the effects of fluctuations. In studying the critical behavior driven by the critical modes \mathbf{a} and \mathbf{b} , one may replace the uniform mode \mathbf{m} by its average value, $\mathbf{m}=(m,0)$, since \mathbf{m} is a noncritical mode induced by the applied magnetic field. Assuming $\mathbf{m} \propto \mathbf{H}$, one arrives at the following reduced Hamiltonian, which will serve as the basis of the analysis:

$$\mathcal{H} = (\nabla \mathbf{a})^2 + (\nabla \mathbf{b})^2 + r(\mathbf{a}^2 + \mathbf{b}^2) + u(\mathbf{a}^2 + \mathbf{b}^2)^2 + v[(\mathbf{a} \cdot \mathbf{b})^2 - \mathbf{a}^2 \mathbf{b}^2] + g(a_x^2 + b_x^2 - a_y^2 - b_y^2) + [\text{terms not containing } \mathbf{a} \text{ and } \mathbf{b}], \quad (5)$$

where the temperature-like variable r and the “effective anisotropy” g are given by

$$r = r_0 + (e + \frac{1}{2}f)m^2, \quad g = \frac{1}{2}fm^2. \quad (6)$$

Let us first investigate the criticality along the two phase boundaries at finite fields. At the (P-II) boundary, the components perpendicular to the applied field, a_y and b_y , become critical since g is positive. Retaining only the critical modes in (5), one easily sees that the resulting GLW Hamiltonian is that of the conventional isotropic $n=2$ ϕ^4 model with $\phi = (a_y, b_y)$ and with the shifted temperature-like variable $\tilde{r} = r - g$. Since $e < 0$ for CsMnBr₃, $\tilde{r} < r_0$ and this line increases in temperature with increasing magnetic field intensity. Thus, the criticality along the (P-II) phase boundary should be of the conventional XY universality. In the intermediate phase, the ordering is such that $a_y \neq 0$ with $a_x = b_x = b_y = 0$ (or other equivalent configurations), and the component parallel to the applied field b_x becomes critical at the (II-I) phase boundary. In deriving the associated GLW Hamiltonian, one may regard a_y given by the mean-field result $a_y^2 = -\tilde{r}/(2u)$, which states that the coefficient of b_y^2 is identical to zero in the intermediate phase. The resulting GLW Hamiltonian is the conventional $n=1$ ϕ^4 model (Ising model) with $\phi = b_x$ and

$$\tilde{r} = r + g + (2u - v)a_y^2.$$

Therefore, the criticality along the (II-I) phase boundary is that of the Ising universality.

The XY and Ising lines merge at $H=0$ giving rise to a novel type of tetracritical point at $T = T_N$. The appropriate GLW Hamiltonian describing the tetracritical behavior is a “chiral Hamiltonian,” \mathcal{H}_0 , whose critical properties were recently studied in detail by the use of renormalization-group techniques for generalized n -component fields $\mathbf{a} = (a_1, \dots, a_n)$ and $\mathbf{b} = (b_1, \dots, b_n)$.^{9(b)} Thus, the associated critical behavior was found to be governed by a new type of $n=2$ chiral fixed point characterized by a set of critical exponents that differed significantly from the standard values: recent numerical estimates based on Monte Carlo calculations give $\alpha \approx 0.40$, $\beta \approx 0.25$, $\gamma \approx 1.1$, and $\nu \approx 0.53$.^{9(c)} Furthermore, it was found that the anisotropy field g , entering the Hamiltonian as in (5), constitutes a scaling field at the chiral fixed point, characterized by the associated anisotropy-crossover exponent ϕ .^{9(b),13} One then has the standard scaling relation for the singular part of the free energy around a tetracritical point,

$$f_s \approx |t|^{2-\alpha} Y \left[\frac{\tilde{g}}{|\tilde{t}|^\phi} \right], \quad (7)$$

where \tilde{t} and \tilde{g} are linear scaling fields,

$$\tilde{t} = t + qh^2, \quad \tilde{g} = h^2, \quad (8)$$

with $t = (T - T_N)/T_N$, $h = H/(k_B T_N)$, and $q \propto (e + \frac{1}{2}f)$. The ϵ - and $1/n$ -expansion calculations^{9(b)} suggest that $1 < \phi < \gamma \approx 1.1$, so that ϕ is probably fairly close to unity. Indeed, a recent numerical estimate based on the two-loop renormalization-group calculation gives $\phi \approx 1.04$.¹⁴

One can derive various observable predictions directly from (7) and (8).¹⁵ (a) The uniform susceptibility for $H=0$ behaves as

$$\chi \sim t^{-\tilde{\gamma}} (1 + cqt^{\phi-1})$$

with

$$\tilde{\gamma} \equiv -(2 - \alpha - \phi) \sim -0.56.$$

Here, we tentatively put $\phi \approx 1.04$ for a rough numerical estimate. The correction term arises because of the fact that the thermal scaling axis, that is the $\tilde{t}=0$ axis, does not agree with the $t=0$ axis. (b) On the thermal axis $\tilde{t}=0$, the magnetization varies as

$$m \sim h^{1/\delta} (1 + cqh^{2(\phi-1)/\phi})$$

with

$$\delta = \phi / (4 - 2\alpha - \phi) \approx 0.48.$$

(c) The two critical lines near the tetracritical point vary as

$$t \sim -qh^2 \pm (h^2/w_\alpha)^{1/\phi},$$

or equivalently, as

$$h^2 \sim w_\alpha |t|^\phi (1 + c_{1\alpha} |t|^{\phi-1} + c_{2\alpha} |t|^{2(\phi-1)} + \dots), \quad (9)$$

where $c_{1\alpha} = \phi w_\alpha q$ and

$$c_{2\alpha} = \frac{1}{2} \phi (3\phi - 1) (qw_\alpha)^2,$$

and $\alpha=1,2$ refers to the (P-II) and (II-I) phase boundaries, respectively.

Note that both critical lines in a close vicinity of the tetracritical point are scaled by a common exponent ϕ . This is to be contrasted to the case of ordinary tetracritical point previously analyzed by Bruce and Aharony¹⁶ and by Mukamel.¹⁷ In such cases the upper critical line was scaled by an anisotropy-crossover exponent ϕ , while the lower critical line was scaled by a slightly different exponent $\phi - \phi_v$, where $\phi_v < 0$ was associated with an irrelevant symmetry-breaking quartic perturbation arising from crystalline anisotropy. Such difference comes from the fact that, in the present case, the chiral fixed point lies at $v^* > 0$, while in the previous cases, symmetry was dynamically restored at the transition and the fixed point governing the tetracriticality was the ordinary XY or Heisenberg fixed point at which $v^* = 0$.¹⁶ Note that, as the fixed point is approached $v \rightarrow v^*$, the lower critical line persists for $v^* \neq 0$, whereas it should disappear for

$v^*=0$. The situation here is analogous to the case governed by the *cubic fixed point* analyzed by Bruce and Aharony.¹⁶

In concluding this section, we add one comment. As is apparent from (9), the nonuniversal correction terms could be significant since ϕ is so close to unity. Therefore, in practice, it might be difficult to estimate the leading exponent ϕ from the experimental data *unless* one has the knowledge of the thermal scaling axis, as was pointed out by Fisher for the case of the bicritical point.¹⁸

III. SCALING ANALYSIS OF CsNiCl₃

In this section, we analyze the axial triangular antiferromagnet CsNiCl₃. The magnetic field is assumed to be applied along an easy c axis as $\mathbf{H}=(0,0,H)$. The appropriate GLW Hamiltonian is given by⁴

$$\mathcal{H}=\mathcal{H}_0+\mathcal{H}_m+\mathcal{H}_A, \quad \mathcal{H}_A=-\delta(a_z^2+b_z^2)-\delta_m m_z^2, \quad (10)$$

where \mathcal{H}_0 and \mathcal{H}_m have been defined by (2) and (3), respectively, but \mathbf{a} , \mathbf{b} , and \mathbf{m} are now three-component vec-

tors. The last term \mathcal{H}_A represents an easy-axis-type anisotropy.

Plumer, Hood, and Caillé⁴ applied the Landau-type free-energy analysis to the Hamiltonian (10), and successfully reproduced the experimentally observed magnetic phase diagram of CsNiCl₃,³ shown schematically in Fig. 1, including a novel type of multicritical point at (T_m, H_m) . According to their analysis, the intermediate phase at lower fields (phase II in Fig. 1) corresponds to the linearly polarized state with $\mathbf{a}\parallel\mathbf{H}$ and $\mathbf{b}=0$ (or equivalently, $\mathbf{b}\parallel H$ and $\mathbf{a}=0$), while the low-temperature phase (phase I) corresponds to the elliptically polarized state with $\mathbf{a}\perp\mathbf{b}$, $|\mathbf{a}|\neq|\mathbf{b}|$, and $\mathbf{a}\parallel\mathbf{H}$ (or $\mathbf{b}\parallel\mathbf{H}$). At higher fields, on the other hand, the ordered state is a 120° structure in the basal plane with the induced magnetization along the c axis, corresponding to $\mathbf{a}\perp\mathbf{b}$, $|\mathbf{a}|=|\mathbf{b}|$, $\mathbf{a}\perp\mathbf{H}$, and $\mathbf{b}\perp\mathbf{H}$ (phase III). The transition between the phases I and III is of first order (spin-flop line), whereas the transitions along all other phase boundaries are continuous.

In order to study the associated critical phenomena, one may derive the following reduced Hamiltonian via the same procedure as was done for the case of CsMnBr₃:

$$\mathcal{H}=(\nabla\mathbf{a})^2+(\nabla\mathbf{b})^2+r(\mathbf{a}^2+\mathbf{b}^2)+u(\mathbf{a}^2+\mathbf{b}^2)^2+v[(\mathbf{a}\cdot\mathbf{b})^2-\mathbf{a}^2\mathbf{b}^2]$$

$$-g(a_x^2+b_x^2+a_y^2+b_y^2-2a_z^2-2b_z^2)+[\text{terms not containing } \mathbf{a} \text{ and } \mathbf{b}], \quad (11)$$

$$r=r_0-\frac{1}{3}\delta+(e+\frac{1}{3}f)m^2, \quad g=\frac{1}{3}(fm^2-\delta). \quad (12)$$

From (11), one can see that the point $(r=0, g=0)$ corresponds to the multicritical point at which the system recovers a full isotropy in spin space, although it is anisotropic in zero field.

The criticality along the three second-order transition lines emanating from the multicritical point, corresponding to nonzero g , may be examined by retaining the associated critical modes in (11), as has been done for CsMnBr₃. Thus, one obtains (i) for the (P-II) phase boundary, $n=2$ ϕ^4 model with $\phi=(a_z, b_z)$ and with the shifted temperature-like variable $\bar{r}=r+2g$; (ii) for the (II-I) phase boundary, $n=2$ ϕ^4 model with $\phi=(b_x, b_y)$ and $\bar{r}=r+g+(2u-v)a_z^2$, on assuming that the symmetry breaking in phase II is such that $a_z\neq 0$; (iii) for the (P-III) phase boundary, $n=2$ chiral model defined by Eq. (2) with $\mathbf{a}=(a_x, a_y)$, $\mathbf{b}=(b_x, b_y)$, and $\bar{r}=r-g$. Therefore, the criticality along the two phase boundaries at lower fields ($H < H_m$) are both XY like. On the other hand, we predict that the criticality along the phase boundary at higher fields ($H > H_m$) is of $n=2$ chiral universality characterized by novel exponents quoted earlier.⁹

The two XY lines and one $n=2$ chiral line meet at a novel type of multicritical point. On the locus $g=0$, which should be tangential to the first-order spin-flop line at the multicritical point, one has the $n=3$ chiral Hamiltonian as is evident from (11). According to the renormalization-group analysis,^{9(b)} its criticality is expected to be governed by a new type of $n=3$ *chiral fixed point*: recent numerical estimates of the associated critical exponents are $\alpha\approx 0.34$, $\beta\approx 0.28$, $\gamma\approx 1.1$, and $\nu\approx 0.55$.^{9(c)}

As g was found to be a scaling field at the chiral fixed point, one again has the scaling relation (7) with linear scaling fields,

$$\tilde{t}=t+qh, \quad \tilde{g}=h-pt, \quad (13)$$

with $t=(T-T_m)/T_m$ and $h=(H-H_m)/(k_B T_m)$, and p is equal to a limiting tangent of the spin-flop line at the multicritical point, $p=(dh_{s-f}/dt)_m$. The exponents appearing in (7), α and ϕ , are now associated with the $n=3$ chiral fixed point. Again, ϕ is expected to be fairly close to unity: a recent numerical estimate based on the two-loop renormalization-group calculation gives $\phi\approx 1.06$.¹⁴

As in the case of CsMnBr₃, various predictions follow directly from (7) and (13). (A) The sublattice magnetization m_s and the associated sublattice susceptibility χ_s on the locus $\tilde{g}=0$ varies as $m_s\sim|t|^\beta$ and $\chi_s\sim t^{-\gamma}$ with appropriate $n=3$ chiral exponents. (B) The discontinuity of the magnetization across the spin-flop line below T_m varies as

$$\Delta M\sim|t|^{\tilde{\beta}}(1+cq|t|^{\phi-1})$$

with

$$\tilde{\beta}\equiv 2-\alpha-\phi\approx 0.60:$$

here we tentatively put $\phi\approx 1.06$ for a rough numerical estimate. (C) The uniform susceptibility χ on the locus $\tilde{g}=0$ diverges as

$$\chi\sim t^{-\gamma}(1+cqt^{\phi-1}+c'q^2t^{2(\phi-1)})$$

with

$$\bar{\gamma} \equiv 2\phi + \alpha - 2 \simeq 0.46 .$$

(D) On the thermal axis, $\tilde{t}=0$, the magnetization deviation $\delta M \equiv M - M_m$ varies as

$$\delta M \sim |h|^{1/\delta} (1 + cq|h|^{(\phi-1)/\phi})$$

with

$$\bar{\delta} \equiv \phi / (2 - \alpha - \phi) \simeq 1.77 .$$

(E) The three critical lines near the multicritical point vary as $g/|t|^\phi = W_\alpha$, where $\alpha = 1, 2, 3$ refers to each phase boundary. All three phase boundaries come in tangent to the first-order spin- ϕ line.

IV. DISCUSSIONS

In this section, we examine our results in the light of the existing experimental data for CsMnBr₃ and CsNiCl₃, and also try to give suggestions concerning the future experiments. Recently, following the theoretical predictions of the possible existence of a new universality,⁹ the criticality of the zero-field transition of CsMnBr₃ was independently investigated by the two groups: Ajiro, Kadowaki, and co-workers gave $\beta \simeq 0.25$, $\gamma \simeq 1.10$, and $\nu \simeq 0.57$ (Ref. 5), while Mason, Gaulin, and Collins gave $\beta \simeq 0.21$, $\gamma \simeq 1.01$, and $\nu \simeq 0.54$.⁶ Both results seem to indicate that the $n=2$ chiral universality is actually governing the tetracriticality of CsMnBr₃. The crossover exponents describing the behavior of the two critical lines around the zero-field tetracritical point have recently been measured by Gaulin *et al.* by means of neutron scattering, giving $\psi_{P-II} \simeq 1.21$ and $\psi_{II-I} \simeq 0.75$.⁷ These experimental values appear to deviate considerably from our present result, $\psi_{P-II} = \psi_{II-I} \simeq 1.04$. The reason for this discrepancy is presently not clear. The existence of a rather large correction to the leading singularity might be the cause, as discussed in Sec. III. Another explanation, suggested by the observation that ψ_{II-I} appears to be smaller than ψ_{P-II} , may be that the *unstable* ($\phi_v > 0$) four-component Heisenberg fixed point lying at $g^* = v^* = 0$ [Ref. 9(b)] is responsible for the observed behavior, since one generally expects $\psi_{II-I} = \phi - \phi_v$ for the Heisenberg-like fixed point with $v^* = 0$.¹⁶ Note that, in this explanation, the observed exponents should be *effective exponents* observable only in some appropriate region in the phase diagram not very close to the tetracritical point. Sufficiently close to T_N , the stable chiral fixed point lying at $g^* = 0$ and $v^* \neq 0$ should describe the asymptotic behavior of the phase boundaries. This scenario could well apply if v is sufficiently small. Meanwhile, if v is small, one may naturally expect that a similar crossover should be observable in the *zero-field* transition. However, indication of such $n=4$ Heisenberg to $n=2$ chiral crossover has *not* been observed in zero field: Indeed, over a rather wide temperature range ($10^{-3} < t < 10^{-1}$), the neutron scattering data of the sublattice magnetization and the corresponding sublattice

susceptibility could be well fitted with a single exponent (β or γ), which is consistent with the $n=2$ chiral value but largely different from the $n=4$ Heisenberg value.⁵⁻⁷ Furthermore, the fact that the theoretical estimate¹⁴ of ϕ_v associated with the $n=4$ Heisenberg fixed point obtained from the ϵ expansion at $O(\epsilon^2)$, $\phi_v \simeq 0.08$, appears to be too small to explain the observed deviation $\phi_v = \psi_{P-II} - \psi_{II-I} \simeq 0.46$ also casts some doubt on the validity of such an explanation. Thus, further careful experimental studies in a close vicinity of the tetracritical point is highly desirable to clarify the situation.

We have predicted that the zero-field uniform susceptibility of CsMnBr₃ should exhibit a singularity of the form $\chi \sim |t|^{-\bar{\gamma}}$ with

$$\bar{\gamma} = -(2 - \alpha - \phi) \simeq -0.56 .$$

This prediction should be testable by straightforward susceptibility measurements.

Concerning the criticality of CsMnBr₃ under finite fields, we have predicted that the upper and the lower transitions should belong to the XY ($n=2$) and the Ising ($n=1$) universality, respectively. Gaulin *et al.* also measured the exponent β at an upper transition point in a finite field ($H \sim 4T$) and obtained $\beta \simeq 0.29$.⁷ Although this value is clearly larger than the corresponding value obtained from the zero-field measurement $\beta \simeq 0.24$, it is substantially smaller than our theoretical value $\beta \simeq 0.35$ for the XY universality. This deviation could possibly be understood as a crossover phenomenon from the $n=2$ chiral behavior to the XY behavior, which is expected for the case of weak magnetic fields.

Next, we turn to CsNiCl₃. Successive phase transitions of CsNiCl₃ in zero field has been studied extensively. As to their criticalities, Clark and Moulton obtained $\beta_{\parallel} \sim \beta_{\perp} = 0.32 \pm 0.03$ by means of the NMR measurement,¹ while Kadowaki, Ubukoshi, and Hirakawa obtained $\beta_{\parallel} \sim \beta_{\perp} = 0.30 \pm 0.02$ by means of neutron scattering,² where β_{\parallel} and β_{\perp} refer to the order-parameter exponent at $T = T_{N1}$ and T_{N2} , respectively. These results seem consistent with the theoretical expectation that the two critical lines at lower fields should belong to the conventional XY universality. The fact that the measured value of β tends to be a bit smaller than the pure XY value $\beta \simeq 0.35$, may be understood as a smooth crossover effect from the $n=3$ chiral behavior, which is realized at the multicritical point.

Very recently, the behavior of phase boundaries near the multicritical point of CsNiCl₃ have been studied by Poirier *et al.* using ultrasonic velocity measurements.¹⁹ These authors have found that the crossover exponents associated with the phase boundaries exceed unity, consistent with the present results. In order to test the theory critically, further careful experimental studies of the critical behavior of CsNiCl₃ are desirable, particularly at the multicritical point as well as along the high-field phase boundary.

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