Chiral critical lines of stacked triangular antiferromagnets under magnetic fields

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The critical behavior of weakly anisotropic stacked triangular antiferromagnets in applied magnetic fields is studied. Under appropriate conditions, an unusual chiral criticality is shown to occur *universally along a critical line.* Several experiments are proposed that might serve to discriminate between the chiral universality scenario for the phase transition of chiral magnets and the nonuniversality scenario. Critical behavior under an applied stress is also discussed.

Critical properties of a variety of frustrated magnets are often entirely different from those of conventional unfrustrated magnets. On the basis of a symmetry argument and Monte Carlo simulations, I claimed that frustrated n-component-vector antiferromagnets on a $d=3$ dimensional stacked triangular (or simple hexagonal) lattice exhibited unusual critical behavior that seemed to lie in a chiral universality class distinct from the conventional $O(n)$ Heisenberg universality class.[1] Indeed, a recent large-scale Monte Carlo simulation on stacked triangular antiferromagnets gave $\alpha=0.34\pm0.06$, $\beta=0.253\pm0.01$, $\gamma=1.13\pm0.05$, and $\nu=0.54\pm0.02$ for $n=2$, and $\alpha=0.24\pm0.08$, $\beta=0.30\pm0.02$, $\gamma=1.17\pm0.07$, and $\nu=0.59\pm0.02$ for $n=3$, which differed significantly from the conventional XY and Heisenberg values.[2] It was predicted that such unusual critical behavior might be observed experimentally in certain stacked triangular antiferromagnets such as $CsMnBr_3$ and $RbMnBr_3$ ($n=2$),[1(b)] and VCl_2 and VBr_2 ($n=3$).[1(a)] Indeed, subsequent experiments carried out on stacked triangular antiferromagnets $CsMnBr_3$, VCl_2, VBr_2, $RbMnBr_3$, and $RbNiCl_3$ support this theoretical prediction in the sense that the exponents as well as the amplitude ratio close to the predicted values were in fact observed.[3]

Theoretical analysis based on the Landau-Ginzburg-Wilson (LGW) Hamiltonian combined with the renormalization-group (RG) $\epsilon=4-d$ and $1/n$ expansions was also made.[4] An appropriate LGW Hamiltonian has been given by

$$\mathcal{H}=\tfrac{1}{2}\{(\nabla\cdot\mathbf{a})^2+(\nabla\cdot\mathbf{b})^2+r(\mathbf{a}^2+\mathbf{b}^2)+u(\mathbf{a}^2+\mathbf{b}^2)^2+v[(\mathbf{a}\cdot\mathbf{b})^2-\mathbf{a}^2\mathbf{b}^2]\}\ , \tag{1}$$

with $0<v<4u$, where $\mathbf{a}$ and $\mathbf{b}$ are n-component-vector fields representing cosine and sine components of the helical (noncollinear) ordering at wave vectors $\pm\mathbf{Q}$ via

$$\mathbf{S}(\mathbf{r})=\mathbf{a}(\mathbf{r})\cos(\mathbf{Q}\cdot\mathbf{r})+\mathbf{b}(\mathbf{r})\sin(\mathbf{Q}\cdot\mathbf{r})\ .$$

It was demonstrated in Ref. 4 that the LGW Hamiltonian (1) is characterized by the $O(n)\times O(2)$ "chiral" symmetry associated with $O(n)$ spin and $O(2)$ phase rotations. The standard $\epsilon=4-d$ and $1/n$ expansions applied to (1) has revealed an unusual *chiral fixed point*[4,5] that is stable for

$$n>12+4\sqrt{6}-[(36+14\sqrt{6})/3]\epsilon+O(\epsilon^2)$$
$$\simeq 21.8-23.4\epsilon+O(\epsilon^2)\ .$$

It was then argued that this chiral fixed point would probably remain stable for $n=2,3$ and $d=3$ and govern the unusual critical behavior observed in stacked triangular antiferromagnets.[4] It was also argued that the same chiral critical behavior should be realized in helimagnets such as Ho, Dy, and Tb, since the LGW Hamiltonian for these helimagnets has the same form as the one describing stacked triangular antiferromagnets.

Thus, there now seems to be reasonable theoretical as well as experimental evidence for the occurrence of a *chiral universality class* in the phase transition of the so-called "chiral magnets" such as stacked triangular antiferromagnets and helimagnets. It should be mentioned, however, that some unresolved problems are still remaining. Among other things, some of the exponents, mainly α and β, experimentally determined for rare-earth helimagnets Ho, Dy, and Tb deviate significantly from the theoretical values for $n=2$ chiral class, although some other exponents, such as γ and ν, are consistent with the theoretical values. In fact, the experimental situation for rare-earth helimagnets is quite confused in that different authors report significantly different exponent values *even for the same material.*[3] For example, the reported values of the exponent β are scattered in the range 0.2–0.4.

Azaria, Delamotte, and Jolicoeur recently claimed that the phase transition of chiral magnets were actually nonuniversal in the sense that it might be either of first order, classical (mean-field) tricritical characterized by the exponents $\alpha=0$, $\beta=\frac{1}{4}$, $\gamma=1$, and $\nu=\frac{1}{2}$, or of conventional O(4) Heisenberg universality characterized by the exponents $\alpha\sim-0.22$, $\beta\sim0.39$, $\gamma\sim1.47$, and $\nu\sim0.74$, depending on the microscopic parameters of the systems:[6] The proposed "nonuniversality" refers to the possible crossover behavior between these different types of transition behavior.

While those authors have derived the conventional O(4) Heisenberg fixed point by means of a RG $\epsilon=d-2$ expansion for the case of $n=3$ (Heisenberg spins),[6] the very existence of this fixed point was seriously questioned recently. It was argued in Ref. 7 that the O(4) fixed point obtained in Ref. 6 was spurious, arising from the perturbative nature of the $\epsilon = d - 2$ expansion, which entirely missed the crucially important nonperturbative effects associated with the vortex degrees of freedom inherent to this system. Furthermore, there is no obvious theoretical reason to expect the classical tricritical behavior especially for chiral magnets. In fact, in terms of the LGW Hamiltonian (1), the Gaussian fixed point at the origin, $u = v = 0$, responsible for such classical tricritical behavior is strongly *unstable* against both quartic parameters u and v.[4] Therefore, in order to reach such a classical tricritical fixed point, one has to tune two *symmetry-unrelated* microscopic parameters so that both renormalized u and v just happen to vanish together. Such coincidence, however, is highly unlikely, since the "initial values" of u and v derived from the microscopic spin Hamiltonian take on finite values of order unity.[1(c),4] Finally, as recently pointed out in Refs. 8 and 2, apparent "nonuniversal" or "crossoverlike" behavior observed experimentally in some rare-earth metals might be due to a mean-field to $n = 2$ chiral crossover caused by the long-range nature of the Ruderman-Kittel-Kasuya-Yosida interaction inherent to these substances, an origin entirely different from the one envisaged in Ref. 6.

Still, one may suspect that the unusual critical behavior observed experimentally and numerically in chiral magnets might be understandable without invoking a new fixed point, arguing that the systems explored in experiments and Monte Carlo simulations *happened* to be just at, or very close to, the unstable Gaussian fixed point by some yet unidentified coincidence, or maybe, the observed unusual exponents are only "effective exponents" caused by some crossover effect between the standard critical behaviors. Although any sign of such crossover has so far been observed in neither large-scale simulations nor in high-precision experiments for *insulators,* it is clearly desirable to further examine the underlying "universality" of the unusual critical behavior of chiral magnets. If the unusual critical behavior is constantly observed for various chiral magnets, which share the chiral symmetry but have very different microscopic parameters, this would refute the above-mentioned suspicion that some kind of "accident" or nonuniversality is responsible for the unusual critical properties. If the observed unusual critical exponents were associated with the classical tricritical behavior as claimed in Ref. 6, for example, such a change in the nonuniversal parameters would induce either the standard O(4) Heisenberg behavior or the first-order transition. Since the exponents predicted for the chiral universality are very different from those for the O(4) Heisenberg universality, the two scenarios then could be distinguished clearly. In this paper, I propose several experimental ways to examine the universality of the chiral critical behavior, which might serve to examine the chiral universality scenario[1,2,4] versus the nonuniversality scenario.[6]

The most straightforward way would be to take various sorts of chiral magnets and investigate the critical properties from one material to another to see whether the predicted unusual critical behavior is really observed in common. In the case of stacked triangular antiferromagnets, candidate materials may include $CsMnBr_3$, $RbMnBr_3$, $CsVCl_3$, VCl_2, and VBr_2, etc. Although there already exists partial experimental support for the universal nature of the critical behavior in these materials,[3] the number of good candidate materials now available is rather small. Therefore, it is clearly desirable to have some experimentally controllable fields that allow one to vary continuously the microscopic parameters of a given material, while observing the chiral symmetry. In the following, I will show that an external magnetic field applied along an appropriate direction in weakly anisotropic stacked triangular antiferromagnets serves for such purposes. From the standpoint of the chiral universality scenario, several predictions for possible experiments will then be given. One obvious advantage to studying stacked triangular antiferromagnets is that most of these substances are *insulators,* and one need not worry about possible crossover from mean-field behavior caused by the long-range interactions as in the case of rare-earth metals. Below, the cases of axial (Ising-like) and planar (XY-like) anisotropies will be considered separately.

(a) Planar (XY-like) stacked triangular antiferromagnets. As mentioned, the zero-field transition of planar stacked triangular antiferromagnets is expected to belong to the $n = 2$ chiral universality. Recently, Gaulin *et al.* have found, by neutron-scattering measurements, that the application of a magnetic field in the basal plane splits a zero-field transition of $CsMnBr_3$ into two successive transitions,[9] establishing that the zero-field transition of $CsMnBr_3$ corresponds to a tetracritical point in the $H_\perp$-T plane.[10] A scaling theory has demonstrated that the reduction of the high chiral symmetry due to the in-plane field is responsible for such a splitting of the transition.[11] The theory also predicts that the criticality along these two critical lines under finite fields is of an ordinary type, each of the XY and Ising universality classes. So, in this case, chiral criticality is realized only at a special isolated point, $T = T_N$ and $H_\perp = 0$. On the other hand, when a magnetic field is applied along the hard c axis, the phase diagram is expected to be as in Fig. 1(a), with the low-temperature phase having a spin configuration in which the spins lie on a cone.[12] In this case, an applied field does not cause the splitting of the transition or change the underlying chiral symmetry. Thus, a critical line in the presence of $H_\parallel$ is expected to be a *chiral critical line* governed by the $n = 2$ chiral fixed point, the same as in the zero-field case all the way along the critical line.

In terms of the LGW Hamiltonian discussed above, the two quartic couplings (u', v') associated with the $n = 2$ chiral Hamiltonian describing the transition under finite $H_\parallel$ are modified from their zero-field values (u, v) as

$$u' = u + c_1 H_\parallel^2 + \cdots , \quad v' = v + c_2 H_\parallel^2 + \cdots , \qquad (2)$$

through the coupling terms between the order-parameter fields and the uniform field $\mathbf{m}$ of the form $c_1' \mathbf{m}^2(\mathbf{a}^2 + \mathbf{b}^2)^2$ and $c_2' \mathbf{m}^2[(\mathbf{a}\cdot\mathbf{b})^2 - \mathbf{a}^2\mathbf{b}^2]$, etc. Here $\mathbf{a} = (a_x, a_y)$ and $\mathbf{b} = (b_x, b_y)$ represent the order-parameter fields in the basal plane. Thus, the quartic couplings under magnetic fields, u' and v', now depend on the field intensity and can be varied continuously. Note that such a change in quartic coupling constants does not affect the asymptotic

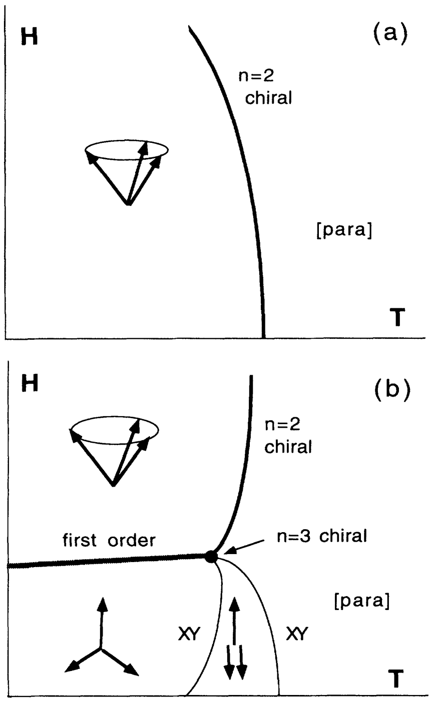

FIG. 1. Magnetic-field-temperature phase diagram of weakly anisotropic, stacked triangular antiferromagnets for each case of planar (XY-like) anisotropy (a), and axial (Ising-like) anisotropy (b). In both cases, the magnetic field is applied along a c axis.

critical behavior so long as the initial values of u and v remain in the domain of attraction of the chiral fixed point, since the chiral fixed point is a stable fixed point. Thus, I predict that the $n=2$ chiral critical behavior should be realized universally along the critical line independent of the field intensity.

On the other hand, if the values of u and v in the zero-field transition of $CsMnBr_3$ happened to lie just at, or very close to, the Gaussian fixed point, as was suggested in Ref. 6, the application of a field along the c axis would immediately deflect the RG flow from the Gaussian fixed point, since the Gaussian fixed point is *strongly unstable* against both u and u perturbations. According to the scenario given in Ref. 6, the asymptotic behavior in such a case should be either a first-order transition or a continuous transition governed by the O(4) Heisenberg fixed point. Therefore, the two scenarios predict entirely different behaviors in the presence of a magnetic field, which can be tested in a straightforward way by suitable experiments.

The exponent α can be measured by standard specific-heat measurements, while the exponents β, γ, and ν can be measured by neutron-scattering measurements. Indeed, recent high-precision specific-heat measurements by Deutchmann *et al.* do indicate that the unusual critical behavior, essentially the same one as in the zero-field case, is realized even under finite fields.[13] Further useful information may be obtained by measuring the uniform magnetization induced by the field. The singular part of this quantity is expected to behave essentially like the energy with the associated singularity, $m_{\parallel}^{\rm sing} \approx |t|^{1-\alpha}$, with $1-\alpha \sim 0.66$ for $n=2$ chiral class. It should be noted that, since the uniform magnetization has a large portion of the regular part induced by the applied field, $m_{\parallel}^{\rm reg} \approx m_0 + m_1 t + \cdots$, care has to be taken in properly subtracting such regular-part contribution in estimating the exponent from the experimental data. The same expression holds also for the zero-field uniform susceptibility when an infinitesimal field is applied along the c axis. By contrast, the singular part of the zero-field susceptibility for an in-plane infinitesimal field is expected to behave as $\chi_{\perp}^{\rm sing} \approx |t|^{2-\alpha-\phi}$ with $2-\alpha-\phi \sim 0.62$ for $n=2$ chiral class, where ϕ is an anisotropy-crossover exponent.[11,14]

(b) Axial (Ising-like) stacked-triangular antiferromagnets. The phase diagram of a weakly anisotropic, axial, stacked triangular antiferromagnet is shown in Fig. 1(b), where a magnetic field is applied along an easy c axis.[12] As can be seen from the figure, three critical lines and one first-order spin-flop line meet at a different type of multicritical point. Kawamura, Caillé, and Plumer examined the critical properties of these systems based on a scaling theory.[11] According to their analysis, the high-field transition line is of $n=2$ chiral universality, while multicriticality at the multicritical point is governed by the $n=3$ chiral fixed point. Then, one can make an independent check of the universality of chiral critical behavior by studying the critical properties along the high-field critical line of an *axial,* stacked triangular antiferromagnet, such as $CsNiCl_3$, $CsNiBr_3$, and $CsMnI_3$. Again, along the high-field critical line, the critical properties should be those of the $n=2$ chiral class, independent of the field intensity. As in the planar case, the singular part of the uniform magnetization should behave as $m_{\parallel}^{\rm sing} \approx |t|^{1-\alpha} \approx |t|^{0.66}$. Note that this singularity is much stronger than the ones along the XY critical lines at lower fields, where one has $m_{\parallel}^{\rm sing} \approx |t|^{1-\alpha_{XY}} \approx |t|^{1.01}$. Note also that, since one has an $n=3$ chiral criticality at the multicritical point, $n=3$ chiral to $n=2$ chiral crossover might occur in the vicinity of the multicritical point along the high-field critical line.

Finally, I note that the application of magnetic fields is not the only way to realize a chiral critical line. One oth-

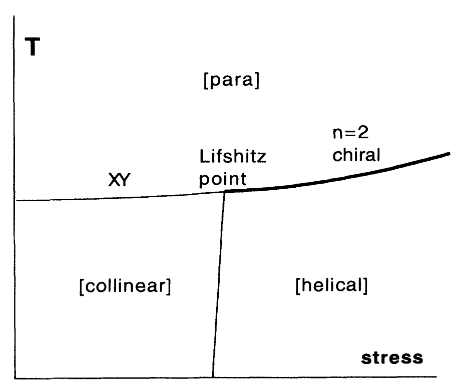

FIG. 2. Schematic phase diagram of an XY-like stacked triangular antiferromagnet under a stress applied in the basal plane.

er possibility may be to apply a uniaxial stress along an appropriate direction in stacked triangular antiferromagnets or helimagnets. Let us consider, for example, an XY-like, stacked triangular antiferromagnet. If one applies an uniaxial stress in the basal XY plane, an *incommensurate* spin structure arises with the associated **Q** vector depending on the stress intensity.[15,16] In this case, the phase diagram is expected to be as given in Fig. 2. The criticality along the paramagnetic-to-helical transition line is of the $n=2$ chiral class irrespective of the **Q** vector or the stress intensity. This chiral critical line ends at a special multicritical point, the so-called Lifshitz point, and the criticality along the other side of the transition line between the paramagnetic and the collinearly ordered phase is expected to be of an ordinary $n=2$ XY criticality. Thus, near the Lifshitz point, a crossover from the Lifshitz-point behavior characterized by the exponents $\beta \sim 0.20$ and $\gamma \sim 1.5$ to the asymptotic $n=2$ chiral behavior might occur. In any case, experimental studies of the critical properties of stacked triangular antiferromagnets, such as $CsMnBr_3$, under an applied stress is of much interest as another experimental check of the universality of chiral criticality.

In summary, I have proposed several possible experiments to examine the chiral universality scenario for the phase transition of chiral magnets versus the nonuniversality scenario. I hope this work will stimulate further experimental activities on related materials.

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[10]It was suggested in Ref. 9 that the tetracriticality observed in $CsMnBr_3$ may oppose to the chiral universality scenario. In fact, this is not necessarily the case: The observed tetracriticality is entirely consistent with, and is rather supportive of, chiral universality scenario as discussed in detail in Ref. 11. It could happen, in principle, that, even at the multicritical point governed by an "ordinary" fixed point, some of the exponents apparently take unusual values when certain relevant scaling fields such as the axial (quadratic) anisotropy mixes into the physical temperature. In such a case, an anisotropy-crossover exponent ϕ comes into play. However, this clearly does not happen in the zero-field transition of $CsMnBr_3$, since symmetry dictates that the effective axial anisotropy always vanishes in zero field and the physical temperature is a *linear* scaling field in itself.

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