

that inhomogeneities can also produce a temperature dependence in the line of maxima for the incipient-triple-point case. Thus, although the triple-point interpretation is still a possibility, the incipient-triple-point interpretation provides an attractive alternative which does not assume the existence of the unobserved fluid-fluid coexistence region.

The triple-point interpretation suggested for the N_2 -graphite phase diagram⁸ also faces the problem of an unobserved fluid-fluid coexistence region. Although no isotherm studies have been made for N_2 in the temperature range of interest, heat-capacity studies^{9,11} have shown anomalies similar to those presented here for krypton. It is known from diffraction probes that both the Kr-graphite system^{4,5} and the N_2 -graphite system^{12,13} have a registered phase. Furthermore, from bulk virial-coefficient studies, the Lennard-Jones (LJ) hard-core parameters are very nearly the same with $\sigma(\text{Kr})=3.60$ and $\sigma(N_2)=3.698$. To the extent that Kr and N_2 behave like the ideal LJ adsorbate, one might expect the phase diagrams for these systems to have essentially identical coverage dependence with a temperature dependence that scales with the ratio of LJ potential strengths $\epsilon(\text{Kr})/\epsilon(N_2)=1.799$. In Fig. 1, the locus of heat-capacity anomalies in the submonolayer regime of the N_2 -Grafoil system is plotted with the temperatures rescaled by $\epsilon(\text{Kr})/\epsilon(N_2)$. The identity of the scaled lines of anomalies establishes experimentally a quantitative correspondence between the Kr and N_2 submonolayer phase diagrams. Near the monolayer the temperature scaling breaks down (Fig. 1). The qualitative correspondence continues to higher coverage, however, and the Kr heat-capacity signa-

ture near the monolayer [Fig. 3(c)] is essentially identical to that previously observed for N_2 in the same coverage range.¹¹ This detailed correspondence between the N_2 and Kr systems suggests that the incipient-triple-point interpretation may also be applicable to the N_2 system.

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Virtual Bicritical Point in CsMnF_3

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The ordering temperature T_c of the easy-plane antiferromagnet CsMnF_3 was measured as a function of magnetic field H . When \vec{H} is perpendicular to the easy plane, T_c decreases monotonically with increasing H , but the decrease is not proportional to H^2 . The latter behavior is explained in terms of a virtual bicritical point which exists mathematically at a negative value of H^2 .

In this Letter we introduce the concept of the virtual bicritical point in easy-plane antiferromagnets, and present experimental data in

CsMnF_3 which support this concept.

The bicritical point (BP) of a low-anisotropy easy-axis antiferromagnet was discussed theoret-

ically in joint papers by Fisher, Nelson, and Kosterlitz (FNK).¹⁻³ Briefly, the application of a magnetic field \vec{H} to an antiferromagnet favors configurations with the staggered magnetization \vec{L} perpendicular to \vec{H} . If \vec{H} is parallel to the easy axis of the antiferromagnet then the H -induced effective easy-plane anisotropy competes with the intrinsic crystalline easy-axis anisotropy. The intrinsic anisotropy dominates at low H , but not at high H . Thus, there are two ordered phases with different directions of \vec{L} : the antiferromagnetic (AF) phase at low H , and the spin-flop (SF) phase at high H . Each of these phases is separated from the paramagnetic (P) phase by a critical line in the temperature-field (T - H) plane. The two critical lines meet at the BP ($T = T_b$, $H = H_b$), as shown in Fig. 1(a). FNK showed that as the critical point $T_c(H)$ moves away from the BP, a crossover due to a change in spin dimensionality takes place. This crossover is similar to the anisotropy crossover in ferromagnets,^{4,5} and it affects the phase boundaries (T_c vs H) near the BP. That is, the phase boundaries near the BP do not follow the predictions of mean-field theory. Experiments⁶ and Monte Carlo calculations⁷ have confirmed many of the FNK predictions.

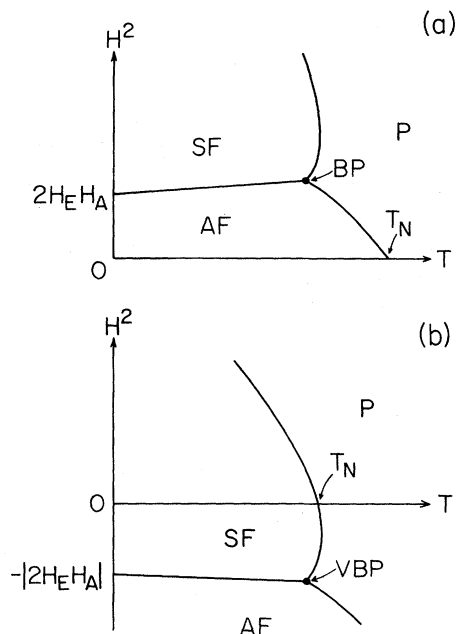


FIG. 1. Schematic of the phase boundaries, in the T - H^2 plane, for (a) a uniaxial easy-axis antiferromagnet and for (b) a uniaxial easy-plane antiferromagnet. \vec{H} is parallel to the symmetry axis. Note that the virtual bicritical point (VBP) in (b) is below the observable boundary (for $H^2 \geq 0$).

In this Letter we consider an easy-plane antiferromagnet with no anisotropy in the easy plane. The transition at the Néel point ($T = T_N$, $H = 0$) is then XY-like. If \vec{H} is applied in the easy plane, a unique preferred direction for \vec{L} is created and the transition becomes Ising-like. The XY-to-Ising crossover, caused by H , should then lead to a bow-shaped phase boundary in the T - H plane.^{3,8} On the other hand, if \vec{H} is perpendicular to the easy plane, then no particular direction in that plane is singled out, and the transition remains XY-like. Because there is no crossover in this case, one might expect that the dependence of T_c on H will be mean-field-like, i.e., T_c will decrease linearly with H^2 as long as H is small compared with the exchange field $H_E(T = 0)$. However, it will be shown that data in the easy-plane antiferromagnet CsMnF_3 do not agree with this expectation. These data are explained by introducing the virtual bicritical point.

CsMnF_3 is a hexagonal easy-plane antiferromagnet.⁹⁻¹⁵ The early interpretations of the magnetic data were based on a two-sublattice model⁹ which led to the following values at $T = 0$: The exchange field is $H_E = 350$ kOe; the uniaxial anisotropy field, which favors the hexagonal plane, is $H_A = -7.5$ kOe; the anisotropy field in the easy plane is $H_a \leq 1$ Oe. A four-sublattice model¹⁴ leads to comparable values. No net spontaneous ferromagnetic moment (canted moment) has been observed in CsMnF_3 . Evidence for the XY character of the transition at $H = 0$ was presented in Ref. 15.

We determined the phase boundaries of CsMnF_3 by measuring the lambda anomaly in the thermal expansion coefficient at different fixed values of H , up to 120 kOe. The experimental procedures were similar to those in Ref. 16. Two single crystals were used. The Néel temperature for both samples was $T_N = 51.38 \pm 0.02$ K. The dependence of T_c on H was measured for two field directions: (1) $\vec{H} \parallel [0001]$, i.e., perpendicular to the easy plane, and (2) $\vec{H} \parallel [10\bar{1}0]$, i.e., in the easy plane. Data for $\vec{H} \parallel [10\bar{1}0]$ were taken in only one sample. The results, plotted in the T - H^2 plane, are shown in Fig. 2. These data are corrected for the demagnetizing field. We focus on the boundary for $\vec{H} \parallel [0001]$. Figure 2 indicates that this boundary is not a straight line, so that the prediction which is based on mean-field theory is incorrect.¹⁷ The interpretation of the boundary for $\vec{H} \parallel [0001]$ is based on a formal similarity between a uniaxial easy-axis and a uniaxial easy-plane antiferromagnets. We assume that \vec{H} is

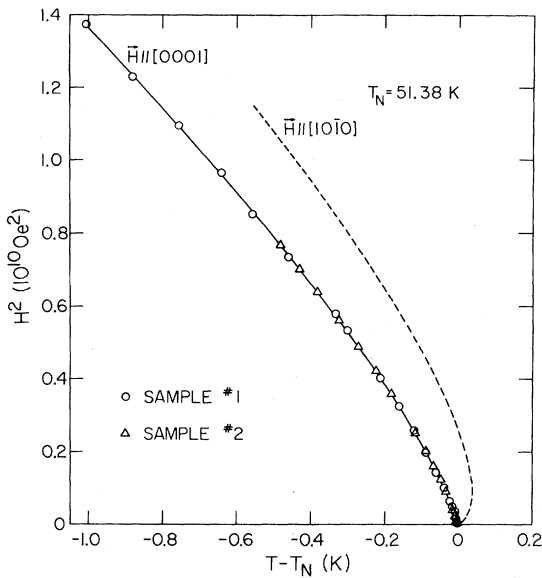


FIG. 2. Measured phase boundaries of CsMnF_3 for $\vec{H} \parallel [0001]$ and $\vec{H} \parallel [10\bar{1}0]$. The data points for $\vec{H} \parallel [10\bar{1}0]$ are detected. The solid line is a least-squares fit to Eq. (1) of the $\vec{H} \parallel [0001]$ data in sample No. 1.

parallel to the symmetry axis. Then, the expressions for the free energies of the easy-axis and the easy-plane antiferromagnets are similar, except for a change of the sign of the uniaxial anisotropy. In the easy-axis case, one obtains the phase diagram shown in Fig. 1(a). To put the easy-plane case on a similar footing, the range of H^2 which appear in the expression for the free energy is allowed to extend beyond the physically allowed range of $H^2 \geq 0$ to the physically forbidden range of negative H^2 . Then, if one proceeds formally, one obtains the phase diagram shown in Fig. 1(b), which is similar to that in Fig. 1(a) except that some of the transitions occur in the experimentally inaccessible range of negative H^2 . We call such transitions virtual transitions. The concept of virtual transitions helps to elucidate the behavior in the physical world of nonnegative H^2 . In particular, the concept of the virtual bicritical point (VBP) explains the departure of the SF-P boundary $T_c(H)$, for positive H^2 , from the mean-field prediction. The effect of the VBP on $T_c(H)$ in the easy-plane case is similar to that of the ordinary BP on the SF-P boundary in the easy-axis case (see Ref. 18). When the magnitude of the easy-plane anisotropy is small, as in CsMnF_3 , the VBP is close to the $H^2 = 0$ axis, and the effect of the VBP on the measured phase boundary $T_c(H)$ is pronounced. That is, the bound-

ary $T_c(H)$ is pronounced. That is, the boundary T_c vs H deviates significantly from mean-field behavior.

To discuss the VBP more explicitly we use the renormalization-group treatment of FNK, but change the sign of $D(\vec{R})$ in Eq. (2.1) of Ref. 3 from that for an easy-axis antiferromagnet to that for an easy-plane antiferromagnet. With \vec{H} parallel to the symmetry axis, the reduced Hamiltonian is still given by Eqs. (3.7)–(3.10) of Ref. 3. However, for a small easy-plane anisotropy, $T_0^\perp > T_0^\parallel$ and a_\parallel is slightly larger than or equal to a_\perp , where the notation is that of Ref. 3. Under these conditions, it is r_\perp which first becomes negative when T is reduced in the presence of a positive (or zero) h_\parallel^2 . Then, one can perform a renormalization iteration in which r_\perp is small and is slowly varying, but r_\parallel is increasing rapidly. This implies an XY-like transition for $H^2 \geq 0$. Conversely, an Ising-like transition occurs at sufficiently large negative H^2 . For some negative H^2 , r_\perp and r_\parallel can be small and of comparable magnitude. Then, a bicritical-type Heisenberg fixed point can be located. This fixed point characterizes the VBP described above. If $H^2 = H_{vb}^2 = -h_0^2$ at the VBP, then the transition is XY-like for $H^2 > H_{vb}^2$, and Ising-like for $H^2 < H_{vb}^2$.

It has been shown² that near the ordinary BP of an easy-axis antiferromagnet, the symmetry-breaking parameter g , which appears in the extended scaling hypothesis,^{4,5} is proportional to $H^2 - H_b^2$. Similarly, in the easy-plane case, g is proportional to $H^2 - H_{vb}^2 = H^2 + h_0^2$. An expression for the SF-P boundary $T_c(H^2)$ near the VBP was obtained by following the procedure of Ref. 2, in which optimal scaling axes were used. Here, we made the additional assumption (to be justified later for CsMnF_3) that the slope of the AF-SF boundary was negligible. Assuming further that the VBP was close to the $H^2 = 0$ axis (i.e., small easy-plane crystalline anisotropy), the following expression for the observable boundary was obtained by subtracting $T_N = T_c(H = 0)$ from $T_c(H)$,

$$T_c(H) - T_N = A[(H^2 + h_0^2)^{1/\varphi} - h_0^{2/\varphi}] - BH^2, \quad (1)$$

where $\varphi = \varphi(n=3) \cong 1.25$ is the crossover exponent, and A and B are positive constants.

The virtual spin-flop transition (AF-SF transition) of an easy-plane antiferromagnet is obtained by following the standard argument for the spin-flop transition of an easy-axis antiferromagnet,¹⁹ but changing the sign of the anisotropy and allowing for the possibility of negative H^2 . The virtual spin-flop transition at $T = 0$ occurs at $H_{vsf}(0)^2$

$\cong -|2H_E H_A| \equiv -H_1^2$. The ordinary spin-flop transition of an easy-axis antiferromagnet occurs at $H_{sf}(0)^2 \cong 2H_E H_A$, for $T=0$. The concept of a virtual spin-flop transition is useful, for example, in considering the spin waves of an easy-plane antiferromagnet in a (real) magnetic field directed parallel to the symmetry axis. The expressions for these spin waves are identical to those of an easy-axis antiferromagnet in the SF phase, except that H_{sf}^2 is replaced by H_{vsf}^2 (see Turov²⁰). In particular, this recipe is applicable to the expressions for the antiferromagnetic resonance frequencies.

The measured boundary $T_c(H)$ of CsMnF_3 , for $\vec{H} \parallel [0001]$, was fitted to Eq. (1) by the least-squares method, holding φ fixed at $\varphi(n=3) = 1.25$. The fit for sample No. 1 gave $A = 0.974\ 31$, $B = 0.205\ 06$, and $h_0 = 41.625$, where H is in units of kilo-oersteds, and T is in millikelvins. For sample No. 2 we obtained $A = 1.022\ 15$, $B = 0.210\ 61$, and $h_0 = 44.397$. The standard deviations for h_0 are 5.1 kOe for sample No. 1, and 2.5 kOe for sample No. 2. The fit for sample No. 1 is shown as a solid line in Fig. 2.

The values obtained for h_0 are in agreement with the following estimate: Consider Fig. 1. In most easy-axis antiferromagnets (e.g., MnF_2 , Cr_2O_3), H_b is within a factor of two of $H_{sf}(0) \cong (2H_E H_A)^{1/2}$. We expect a similar behavior in most easy-plane antiferromagnets. That is, $H_{vb}^2 \equiv -h_0^2$ should be comparable to $H_{vsf}(0)^2 \cong -|2H_E H_A| \equiv -H_1^2$. In CsMnF_3 , a value $H_1 = 41.1 \pm 0.6$ kOe was deduced from antiferromagnetic resonance measurements.¹³ This value is indeed comparable to our values for h_0 . The closeness of H_1^2 to h_0^2 also implies that the slope of the (virtual) AF-SF line in CsMnF_3 is quite small, i.e., $d(H^2)/dT \lesssim 10^7$ Oe²/K which is small compared to values of $H^2/(T_N - T)$ in Fig. 2. This justifies the neglect of the slope of the AF-SF line in the derivation of Eq. (1).

A discussion of the phase boundary for $\vec{H} \parallel [1010]$, which is bow shaped because of the XY-to-Ising crossover, will be presented elsewhere.

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¹⁸A physical insight into the non-mean-field behavior of $T_c(H)$ may be gained from the following argument. Starting from a fictitious fully isotropic antiferromagnet at $H=0$, the introduction of an effective easy-plane anisotropy, of magnitude g , would lead to a Heisenberg-to-XY crossover. Such a crossover would result in a non-mean-field behavior of T_c vs g . For small g , T_c should increase by an amount proportional to $g^{1/\varphi}$, where $\varphi = \varphi(n=3) \cong 1.25$. In an antiferromagnet with a given easy-plane crystalline anisotropy, the ordering transition at $H=0$ is already XY-like. When a (real) magnetic field is applied parallel to the symmetry axis (i.e., perpendicular to the easy plane), it favors confi-

gurations with \vec{L} in the easy plane. Thus, the effective easy-plane anisotropy g , which at $H=0$ is due solely to the crystalline anisotropy, is enhanced by a magnetic field. The magnetic field, therefore, continues the crossover processes which was initiated by the crystalline anisotropy. The non-mean-field behavior of T_c vs H [which is expressed in Eq. (1) by the term in the

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Nature of the Incommensurate Phase in Ammonium Fluoberyllate

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Elastic scattering of 14.4 keV Mössbauer γ rays from incommensurate ammonium fluoberyllate indicates that the distortion is essentially static and that phasons do not make anomalously large contributions to the Debye-Waller factor of satellite reflections.

Ammonium fluoberyllate (AFB) was shown by Iizumi and Gesi¹ to exist in an incommensurate form between 175 and 181 K. Within this temperature range relatively intense satellite peaks can be observed characterized by the wave vector $\vec{Q} = (0.5 - \delta)\vec{a}^*$, so that the scattering vector of one of these reflections is given by $\vec{k} = \vec{H} \pm \vec{Q}$, where \vec{H} is a vector of the reciprocal lattice existing above 181 K. At 175 K the value of δ drops discontinuously to zero from a value of approximately 0.015.

There is much interest in the crystal dynamics of an incommensurate phase. Overhauser² drew attention to a possible excitation he called a phason that corresponds to phase modulation of the incommensurate distortion. By taking the ground state to be a single plane wave, the atomic displacements may be represented by

$$u(\vec{L}) = A \sin[\vec{Q} \cdot \vec{L} + \varphi(\vec{L}, t)],$$

$$\varphi(\vec{L}, t) = \sum_{\vec{q}} \vec{q} \sin(\vec{q} \cdot \vec{L} - \omega t).$$

Overhauser went on to show that phason deviations from the ground state constitute a coherent superposition of phonon modes with wave vectors $\vec{q} + \vec{Q}$ and $\vec{q} - \vec{Q}$. Long-wavelength phasons, then, are located near satellite peaks in k space. Furthermore in the phason branch $\omega(\vec{q}) \rightarrow 0$ as $\vec{q} \rightarrow 0$. The most remarkable feature of Overhauser's phasons is that each phason contributes to the exponent of the Debye-Waller factor an amount typically 100 times larger than that of a phonon with the same frequency. Consequently the elastically scattered intensity at positions $\vec{H} \pm \vec{Q}$ should be

vanishingly small. But since long-wavelength phasons produce scattering very close to these positions, the total intensity, the sum of elastic and one-phason scattering, could still be substantial when integrated over a small region of k space around $\vec{H} \pm \vec{Q}$; the scattering observed at satellite positions could be all inelastic. While recent experiments³ and theoretical predictions^{4,5} have shown the ground state of an incommensurate phase not to be a single plane wave, thereby casting doubt on the relevance of Overhauser's predictions, it is still clearly important to perform an energy analysis of the scattering at satellite positions.

We have done this in a Mössbauer γ -ray scattering experiment. Electromagnetic radiation is obtained from the decay $^{57}\text{Co} \rightarrow ^{57}\text{Fe}$ which produces γ rays of 14.4 keV. The cobalt atoms are diffused into a rhodium foil, 6 μm thick, of dimensions $5 \times 2 \text{ mm}^2$, with the effect that about 70% of the γ rays are emitted without recoil. The 150-mCi source is held stationary and the radiation scattered by the stationary crystal is passed through an absorber containing ^{57}Fe before detection by the counter. With the absorber also held stationary, nearly all γ rays elastically scattered by the crystal will be resonantly absorbed within the absorber, while γ rays scattered with an energy change a little greater than the linewidth of the absorber will not be resonantly absorbed. By vibrating the absorber one can destroy all resonant absorption, and so from the difference in counting rates obtained with stationary and moving absorbers the elastic and inelas-