Lifshitz Point in MnP

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Measurements of the transverse differential susceptibility in MnP suggest that the para-ferro-fan triple point is a Lifshitz point (LP). At this point all phase boundaries are tangent to each other, and the λ line exhibits an inflection point. These features agree with predictions for a LP characterized by $d=3$, $n=m=1$, which is expected for this crystal symmetry. A crossover exponent $\phi=0.634\pm 0.03$ is obtained, in agreement with theory.

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The Lifshitz multicritical point was introduced theoretically by Hornreich, Luban, ans Shtrikman.[1] In a magnetic system, the Lifshitz point (LP) is a triple point where paramagnetic, ferromagnetic (or antiferromagnetic), and helicoidal phases meet. Thus, the LP divides the λ into two segments: On the first segment, the paramagnetic (disordered) phase transforms into a ferromagnetic phase with local magnetization which is uniform in space, i.e., $\vec{k}=0$. On the second segment, the paramagnetic phase transforms into a helicoidal phase with local magnetization which varies in space, with a wave vector $\vec{k}$ which approaches zero as the LP is approached along the λ line. In this Letter we present experimental evidence for the existence of a LP in MnP; data for the phase boundaries near the LP; and an experimentally derived crossover exponent. To our knowledge, this is the first experimental observation of a LP in a magnetic material.

MnP is a magnetic compound with orthorhombic structure ($a>b>c$) which exhibits several ordered phases.[2-5] In the absence of a magnetic field MnP is ferromagnetic between 47 and 291 K, with moments parallel to the c axis. Below 47 K a screw phase is observed in which the moments rotate in the b-c plane with a propagation vector $\vec{k}$ along a. A second helicoidal phase, called the fan phase, is observed when a magnetic field $\vec{H}$ is applied along the b axis. In the fan phase the moments are in the b-c plane and $\vec{k}\parallel a$, but the moments do not undergo a full rotation in the b-c plane. A rough outline of the phase diagram when $\vec{H}$ is parallel to the b axis, based on earlier works,[2,5] is shown in the inset of Fig. 1. In this Letter we focus on the triple point where the paramagnetic, ferromagnetic, and fan phases meet. Previous works suggest that the ferro-fan transition is of first order; that the para-to-ferro transition is of second order and is associated with the development of a uniform magnetization component along the c axis; and that the para-to-fan transition is of second order and is associated with the development of an oscillatory magnetization component along the c axis. Near both second-order transitions there is a continuous

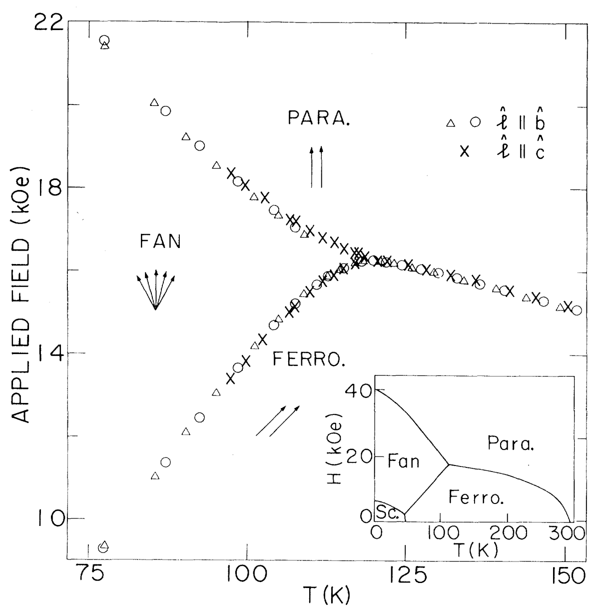

FIG. 1. Phase diagram of MnP near the para-ferro-fan triple point, as determined from magnetostriction measurements. The magnetic field $\vec{H}$ is parallel to the b axis. The unit vector $\hat{l}$ specifies the direction along which the sample's length l was measured. The inset shows the entire phase diagram, for $\vec{H}\parallel b$, based on earlier studies (Refs. 2, 5).

variation of the b component of the magnetization.

We determined the phase boundaries near the para-ferro-fan triple point from mangetostriction (MS) measurements, and from susceptibility measurements. The magnetic field was parallel to the b axis. In the MS measurements, the sample's length l was measured as a function of H at constant temperature T. The data were differentiated numerically to obtain $\partial l/\partial H$. The first-order transition (ferro-fan) appeared as a spike in $\partial l/\partial H$. The second-order transitions (para-ferro and para-fan) appeared as λ anomalies in $\partial l/\partial H$. Figure 1 shows the phase boundaries obtained from MS data on a $0.84\times0.97\times2.67\text{-mm}^3$ sample, with the long dimension along the b axis. Susceptibility data on another sample led to a phase diagram which agreed with Fig. 1.

Near the triple point (T_t,H_t) the shapes of the phase boundaries in Fig. 1 are qualitatively different from those in the inset. All phase boundaries appear to be tangent to each other. The para-fan boundary is concave up. The para-ferro boundary is concave down. Thus the λ line has an inflection point at the triple point. From Fig. 1 we estimate that $T_t=121\pm1$ K, which is more than 10 K higher than values quoted earlier.

We now present transverse susceptibility data which are consistent with the interpretation that the triple point is a Lifshitz point. The transverse differential susceptibility was measured with a steady applied field H_b parallel to b and a small modulation field h_c parallel to c. The measured quantity was $\chi_c=\partial M_c/\partial h_c$ as a function of H_b, where M_c is the net magnetization along the c axis. The basic idea was as follows. When the ferro phase is approached from the high-field side (i.e., H_b decreases toward the phase boundary), the ordering susceptibility χ_c increases, until it diverges at the para-ferro transition. In the ferro phase, χ_c should remain large. On the other hand, at the para-fan transition the (uniform) susceptibility χ_c should not diverge. The susceptibility which should diverge at this transition is $\chi_c(\vec{k}_\lambda)$ for a fictitious modulation field parallel to the c axis but with a periodicity in real space which matches that of the fan phase on the λ line. If the triple point is a LP, then the wave vector $\vec{k}_\lambda$ of the fan phase on the λ line should approach zero when $T\to T_t$. Then $\chi_c(\vec{k}_\lambda)\to\chi_c$ as $T\to T_t$. Therefore, as one moves along the para-fan boundary, the uniform susceptibility χ_c should diverge when T_t is reached. In practice, the measured χ_c does not diverge because it is a derivative with respect to an applied magnetic field rather than with respect to an internal magnetic field. When the intrinsic susceptibility χ_c^{in} (with respect to internal field) becomes very large, the measured χ_c approaches the upper bound $1/N$, where N is the demagnetizing factor. All our data are for the measured χ_c.

Measurements of χ_c were performed on a $2.6\times2.6\times0.9\text{-mm}^3$ single crystal, with the long dimensions parallel to b and c. The modulation field had an amplitude of 4 Oe and a frequency of 188 Hz. Data were taken at fixed T between 88 and 146 K. Some results for χ_c vs H_b are shown in Fig. 2. The data for 125 K are typical for $T>T_t=121$ K. With decreasing H_b, χ_c increases and reaches a nearly constant value when the sample enters the ferro phase. This is the expected behavior for $T>T_t$ except that the measured susceptibility is not perfectly constant in the ferro phase. We attribute this departure from ideal to a small misalignment of the steady field from the

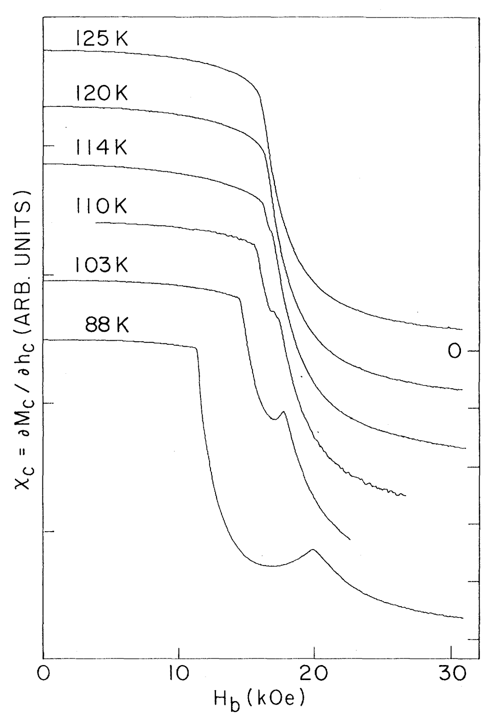

FIG. 2. Transverse differential susceptibility, χ_c, measured with a steady magnetic field H_b along b and a modulation field h_c parallel to c. Each curve shows $\chi_c=\partial M_c/\partial h_c$ (where M_c is the net magnetization along c) as a function of H_b at a fixed T. The curves were displaced in the vertical direction relative to each other. The zeros for successive curves are indicated by short horizontal lines on the right. Apart from this zero shift, the vertical scale is the same for all curves. The values of χ_c at $H=0$ are very nearly equal.

b axis.[6] The curve for $T = 88$ K shows a well-defined peak at a field H_2 where the para-fan transition occurs. In addition, near the fan-ferro transition, at H_1, χ_c increases and reaches a nearly constant value. At 88 K, the susceptibility $\chi_2 \equiv \chi_c(H_2)$ is much smaller than the susceptibility χ_1 in the ferro phase for a field just below H_1. As T increases toward T_t, χ_2 increases and the separation $H_2 - H_1$ decreases. At 103 K, the susceptibility peak at H_2 is still quite distinct, but at 110 K only a "shoulder" is observed at H_2. This "shoulder is still visible at 114 K. Between ~116 K and $T_t = 121$ K the "shoulder" was not resolved, and the curves resembled those at $T > T_t$. It is significant that below ~116 K, $\chi_1 - \chi_2$ decreased with increasing T. The results in Fig. 2 strongly suggest that $\chi_2 \to \chi_1$ when $T \to T_t$, i.e., at T_t the susceptibility of the fan phase equals that in the ferro phase. This conclusion is also supported by a plot (not shown) of $\chi_1 - \chi_2$ vs T.

The intrinsic susceptibility χ_c^{in} is higher than the measured χ_c. The data indicate that as the triple point is approached along the para-fan boundary, χ_c^{in} approaches the very large value of the intrinsic susceptibility on the ferro side of the ferro-fan boundary. This is the expected behavior at a LP. However, the data for χ_c do not prove that the triple point is a LP because a simultaneous divergence (at the triple point) of χ_c and $\chi_c(k_\lambda \neq 0)$ is possible, in which case $\vec{k}$ jumps from a finite value to zero. Evidence that the triple point is a LP is provided by the phase diagram in Fig. 1, as follows.

In an orthorhombic crystal, such as MnP, a ferromagnetic LP should be characterized by a lattice dimensionality $d = 3$, an order parameter with one component ($n = 1$), and a unique direction for the instability wave vector ($m = 1$).[7] In the present case the single-component order parameter is associated with the alignment of the magnetic moments along the c axis. The magnetic moment along b is nonzero both in the disordered and in the ordered phases, and it should not constitute an additional component for the order parameter. The direction of $\vec{k}$ is parallel to a.

All phase boundaries near a LP with $d = 3$, $n = m = 1$, were calculated in mean-field theory.[8] The λ line (but not the first-order line) was also calculated from high-temperature series.[9] Both calculations show that the two segments of the λ line are tangent to each other at the LP. The mean-field calculation also shows that the first-order line is tangent to the λ line at the LP. According to the series calculation, the λ line has an inflection point at the LP. The results in Fig. 1 agree with these predictions.

One difference between Fig. 1 and the theoretical phase diagrams in Refs. 8 and 9 is the choice of coordinate axes. In Fig. 1, T and H_b are the coordinates. In the theoretical papers, T and the pressure p, or T and a certain ratio of the exchange constants, are the coordinates. However, there is no correspondence between the T axis in Fig. 1 and that in the theoretical papers, and the same is true for the second axis. The physical reason for this difference is that in MnP a temperature change (at constant H_b) leads to a change in the ratio of the exchange constants.[10] To achieve a correspondence between the phase diagram in Fig. 1 and that in the theoretical papers it is necessary to use generalized scaling axes in both phase diagrams: One axis is tangent to the λ line at the LP; the second axis may be taken along any other direction, although optimal scaling[11] may dictate a specific direction.

Under the assumption of generalized scaling, the shape of each of the two segments of the λ line near the LP is governed by the crossover exponent ϕ. For $n = m = 1$, the ϵ expansion to first order in ϵ gives $\phi = 0.625$.[1,12] Our attempts to obtain ϕ from the measured λ line were hindered by the fact that the tangent at the LP (which is required in the analysis) could not be determined with sufficient precision. For this reason, an alternative analysis was performed, making use of both the first-order (ferro-fan) line $H_1(T)$ and the para-fan segment $H_2(T)$ of the λ line. We assumed that generalized scaling near the LP was also valid at the first-order line, which implied that the exponent ϕ described the shape of this line also. Such an assumption had been used for the first-order line near a tricritical point.[13] Mean-field calculations[8] show that the exponent describing the first-order line near a LP with $n = m = 1$ is the same as for the λ line. However, we know of no general proof of this conclusion.

We chose one scaling axis X_1 tangent to the λ line at the LP, and another, X_2, parallel to the H_b axis. Near the LP the difference $\Delta H_b \equiv H_2 - H_1 = \Delta X_2$ (for the same T) should be proportional to $X_1^{1/\phi}$, which is proportional to $(T_t - T)^{1/\phi}$. The least-squares fits will be discussed in a later publication. From these fits we estimate that $\phi = 0.634 \pm 0.03$. It is noteworthy that the phase boundaries which were presented and analyzed were not corrected for the demagnetizing field. However, the estimated corrections are small (several hundred oersteds), have an insignificant effect on ϕ, and do not change our other conclusions.

In summary, the data for χ_c, the overall shapes of the phase boundaries, and the experimentally derived ϕ support the conclusion that the para-ferro-fan triple point in MnP is a LP.

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Observation of Solid ^{3}He Ordering at Melting Pressures in High Magnetic Fields

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With use of compressional cooling, ordering in solid ^{3}He is observed and identified by a null in the pressurization rate for reversible compressions in magnetic fields up to 7.2 T. At this field the ordering is above 3 mK. Entropies at T_A deduced from $(P_{A_2} - P_{A_1})/(T_{A_2} - T_{A_1})$ are shown to be lower than expected from the various models. Irreversible magnetic heating at high fields which limits the effective cooling power is also observed.

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A rapid decrease in the entropy of solid ^{3}He at 1.1 mK was first identified as the nuclear ordering transition by Halperin *et al.*[1] The effect of a moderate magnetic field on the ordering has been studied by several groups,[2,3] stimulating a variety of theoretical models: multiple spin exchange,[4] defect-induced[5] or polaron-induced ordering,[6] and spin-glass.[7] In this Letter, we report pressure measurements in magnetic fields up to 7.2 T as well as results of entropy measurements at the temperature 2.75 mK.

The compressional cell,[8] including the compressional membrane, was constructed entirely from plastic. The liquid volume was 2.1 cm^3.

