Evidence for the Ising-Lifshitz crossover in MnP

C. C. Becerra,* V. Bindilatti, and N. F. Oliveira, Jr.
Instituto de Física, Universidade de São Paulo, Caixa Postal 66318, CEP 05315-970 São Paulo, SP, Brazil

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Manganese phosphide (MnP) is one of the few magnetic systems in which a uniaxial Lifshitz point (LP) has been identified. For the external magnetic field applied along the crystallographic b axis, the LP occurs at $T_L = 121 \pm 1$ K and $H_b = 16.4$ kOe, at the confluence of modulated, ferromagnetic (F), and paramagnetic (P) phases. It has been shown that the magnetic susceptibility at fixed temperature (χ) as a function of H_b diverges at the F-P phase boundary with the specific heat exponent α. In this work $\chi \times H_b$ was measured across the F-P phase boundary from $T \approx 200$ K down to $T = 121$ K. From the analysis of the divergence, α was evaluated at different temperatures. For T well above T_L, $\alpha \approx 0.12$, a value close to the prediction for the three-dimensional Ising model. As T is lowered, the divergence gets sharper and α increases, reaching $\alpha = 0.44$ at T_L, in agreement with predictions for this uniaxial LP.

I. INTRODUCTION

Previous studies made of MnP (Refs. 1–3) stressed its connection with the $d=3$ anisotropic next-nearest-neighbor Ising models that show a uniaxial Lifshitz point (LP) in the $T \times \kappa$ plane (T is temperature and $\kappa = J_2/J_1$ is the ratio between competing antiferromagnetic and ferromagnetic exchange interactions). In mean-field theory $\kappa = -0.25$ at the LP. Lifshitz points arise in magnetic systems with competing interactions at the confluence of a modulated and a ferromagnetic phase (F) with the paramagnetic phase (P). The transitions from the ordered phases to the P phase are second order. In MnP, the modulated phase near the observed LP is a fanlike one (FAN),[2] with the wave vector **q** in the a direction (the hard magnetic axis). Neutron-diffraction measurements of spin-wave dispersion curves in MnP,[4] interpreted in terms of a model with interactions J_1 and J_2, respectively, between nearest- and next-nearest-neighbor planes perpendicular to the a axis, showed that κ is T dependent, varying between -0.27 and -0.23 from 70 to 200 K. At the LP temperature (T_L) κ is near -0.25. Thus, in MnP, the variable controlling κ is T and a small interval in κ is mapped in a wide range of T. Since it is naturally tuned close to the critical ratio for the competing interactions, MnP is an ideal material to study the critical behavior near the LP.

MnP has an orthorhombic structure ($a = 5.92$ Å, $b = 5.26$ Å, $c = 3.17$ Å) and becomes ferromagnetic at 291 K, with its magnetic moments aligned along the c axis (easy axis). The phase diagram of MnP obtained when the magnetic field **H** is applied perpendicular to the easy axis shows a line of multicritical LP's. This line of LP occurs at $T \approx 121$ K,[5] and extends from $\mathbf{H} \| a$ to $\mathbf{H} \| b$ (see Fig. 1 of Ref. 5 for the phase diagram in the $H_b \times H_a \times T$ space). We refer also to previous papers for a detailed discussion of the richness of phases and the different types of critical behavior encountered in this material. What concerns us in this paper is the critical behavior along the boundary that delimits the F phase—in particular, the second-order line contained in the $H_b \times T$ plane that ends at the LP. At temperatures much higher than T_L, the critical behavior along this line is expected to be of the Ising type and should crossover to a Lifshitz critical behavior close to the LP.

In Fig. 1, the phase diagram in the $H_c \times H_b \times T$ space is schematized. The portion of this phase diagram involving the FAN phase was experimentally explored in Ref. 2. This diagram is also based on the theoretical study of Ref. 6. For $\mathbf{H} \| c$ the applied field corresponds to the ordering field for the F phase. The surface separating the ↑ and ↓ magnetic moment orientations is a coexistence surface. Transitions across this surface are first order. The coexistence surface is limited for temperatures above T_L by the second-order λ line that separates the F from the P phase. For increasing applied fields along the b axis, a genuine phase transition is only possible if the field is rigorously along b (no c component). We will see how important it is to get rid of any c component of **H** in order to observe the mentioned crossover.

Our analysis of the nature of the critical exponent along the F-P phase boundary will be strongly based on the theoretical paper of Riedel and Wegner[7] on the scaling approach to anisotropic magnetic systems. When an anisotropy axis is present in a ferromagnet and the magnetic field $H_\perp$ is applied

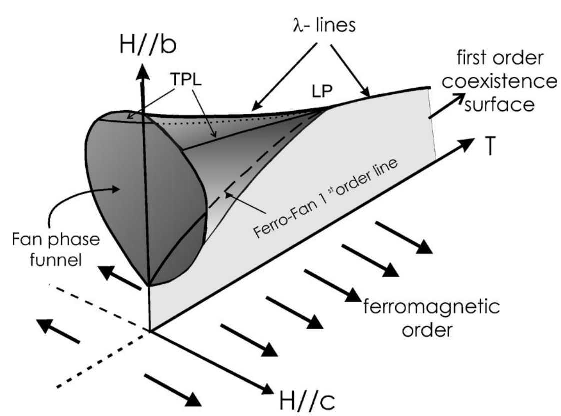

FIG. 1. Schematic phase diagram of MnP close to the Lifshitz point (LP) in the $T \times H_b \times H_c$ space. H_c is the ordering field for the F phase. The surface separating the ↑ and ↓ directions is a coexistence surface which is limited, for $T > T_L$, by the second-order F-P λ line. For $T < T_L$, second-order FAN-P transitions occur across a surface limited by the two lines of tricritical points (TPL).

perpendicular to this direction, the Curie temperature T_C decreases with increasing $H_\perp$ and a line of ordinary critical points is generated in the $T\times H_\perp$ plane. In Fig. 1 this line corresponds to the critical λ line that delimits the coexistence first order surface. Along this line, the critical exponents are those associated with the universality class of the critical Curie point. In MnP this line is expected to be Ising like because the order parameter that corresponds to the magnetization along the c axis has a single component $(n=1)$. In the scaling assumption made by Riedel and Wegner, the thermodynamic functions close to this λ line will depend on $H_\perp - H_\lambda$ in the same way as they depend on $T-T_C$ when $H_\perp=0$. For instance, in their analysis they find that

$$m_\parallel \propto |H_\lambda - H_\perp|^\beta, \quad m_\perp^{\mathrm{sing}} \propto |H_\lambda - H_\perp|^{1-\alpha}, \tag{1}$$

$$\chi_\parallel \propto |H_\lambda - H_\perp|^{-\gamma}, \quad \chi_\perp^{\mathrm{sing}} \propto |H_\lambda - H_\perp|^{-\alpha}, \tag{2}$$

where $m_\parallel$ and $\chi_\parallel$ are the components of the magnetization and the susceptibility measured along the easy axis, and $m_\perp^{\mathrm{sing}}$ and $\chi_\perp^{\mathrm{sing}}$ the singular parts of the magnetization and susceptibility measured along the transverse direction. The indices β, γ, and α are the usual critical exponents associated with the magnetization, susceptibility, and specific heat. The relevant point for us in this work is that the transverse susceptibility diverges along the λ line with the critical exponent α, usually associated with the specific heat. The possibility to tune very precise values of κ by means of the temperature T, together with the fact that $(H_\perp - H_\lambda)$ can be associated with the thermal axis, permits that the measurement of $\chi_\perp = \partial m_\perp / \partial H_\perp$ as a function of $H_\perp$ (the thermal axis) at fixed temperature (that is, at a fixed value of κ) can be used to obtain the value of the exponent α along the F boundary line. What is expected is that, as the LP is approached, the values of the critical exponents will ''cross over'' to the values associated with the Lifshitz universality class. The value of the critical exponent α_L at the LP was already measured, together with the critical amplitude ratio A_+/A_-. The values are $\alpha_L=0.46$ and $A_+/A_-=0.65$.[8] For the Ising model the predicted values for these two critical parameters are $\alpha_I=0.125$ and $A_+/A_-=0.55$.

Hornreich, Luban, and Shtrickman[9] worked out the theoretical predictions for the LP. Two pairs of critical exponents are introduced (ν_2,ν_4) and (η_2,η_4), which describe the critical correlations in the m and $d-m$ subspaces (m is the dimensionality of the wave vector $\mathbf{q}$ of the modulated phase). The upper critical dimension for the Lifshitz behavior is $d_c=4+m/2$. To first order in $\epsilon_L=d-d_c$,

$$\nu_4=\frac{\nu_2}{2}=\frac{1}{4}\left(1+\frac{n+2}{2(n+8)}\epsilon_L\right)+\mathcal{O}(\epsilon_L^2). \tag{3}$$

These critical exponents relate to α_L through the hyperscaling relation

$$2-\alpha_L=m\nu_4+(d-m)\nu_2. \tag{4}$$

For the uniaxial LP point ($d=3$, $n=1$, $m=1$), we have $\epsilon_L=3/2$ and $\alpha_L=7/16=0.4375$. Although the amplitude ratio at LP's has been the object of recent theoretical work,[10,11] no predictions exist for the uniaxial LP in question.

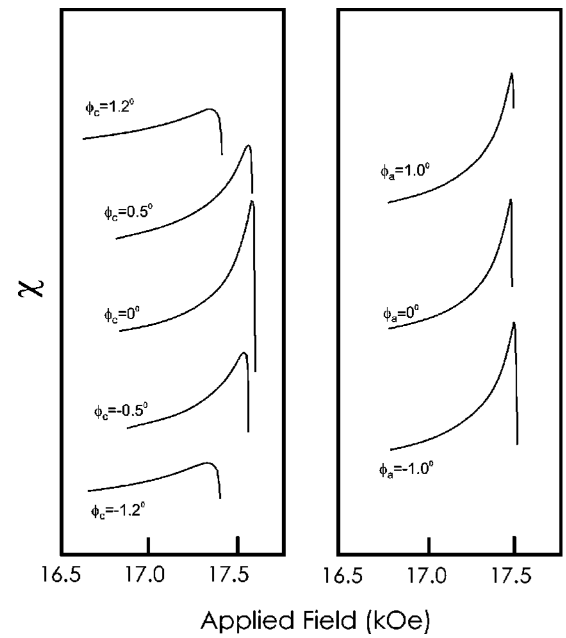

FIG. 2. Experimental curves for $\chi\times H$ at $T=126$ K, obtained in the process of orienting the field along the b axis. The magnet was rotated relative to the sample in the bc plane, in order to eliminate any c component of the applied field, and also in the ba plane. The deviation angle of the field from the b axis is indicated for each trace.

II. EXPERIMENTAL SETUP AND PROCEDURES

Measurements of the ac susceptibility were performed on a MnP single crystal, cut into a 5.2-mm-diam sphere. We used a frequency of 17 Hz and a modulation field $h_{\mathrm{ac}}=2.5$ Oe. The sample was mounted at the end of a rod that fitted into a small Dewar that also accommodates the pickup coils. The temperature T was measured with a carbon-glass resistor in direct contact with the sample. In this setup T could be varied from 1.2 K to room temperature and maintained constant within 0.01 K for long periods of time. The runs were made at fixed T with H varied at a rate of 3.4 Oe/s.

The superconducting magnet was mounted in a platform which can be tipped around the vertical in order to permit a fine alignment of the field axis (within $\pm 4°$). The sample was mounted with its b axis along the axis of the pickup coils and very close to the applied field direction $(\mathbf{H}\| h_{\mathrm{ac}} \| b)$. The final alignment was made *in situ* by gradually tipping the magnet in order to eliminate any component of the field along the c axis (in other words, to eliminate any component of the ordering field for the F phase). The shape of the susceptibility curves at the F-P transition near the LP was used to monitor this procedure. In this experiment the alignment was made at $T=126$ K, but control checks were also made at higher temperatures. In Fig. 2 the shape of the susceptibility curves near the phase boundary is shown as a function of the deviation from the b axis, towards the c axis (ϕ_c) and also towards the a axis (ϕ_a). A component of the field along the c axis tends to destroy the second-order transition. The curves get progressively rounded, and only a 1° misalignment is sufficient to ruin the experiment. The deviation towards the a axis is not critical since we are still crossing a second-order transition surface. This is clearly shown in Fig. 2 as the absence of rounding with respect to the $\phi_a=0$ trace. The final align-

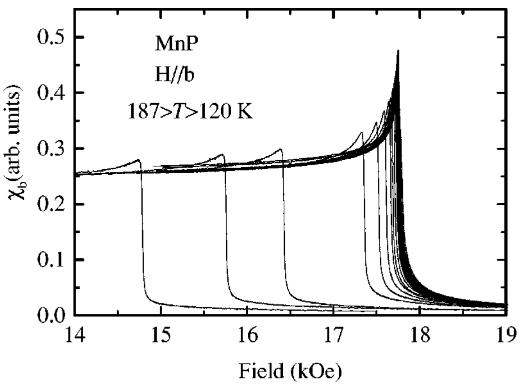

FIG. 3. Isothermal $\chi \times H$ curves with **H** along the b axis. From left to right the temperatures are 187.4, 170.5, 154.0, 132.4 K,... . The the lowest temperature is $T = 120$ K.

ment was estimated to be better than 0.05° in the bc plane and 0.5° in the ab plane.

III. RESULTS AND DISCUSSION

In Fig. 3 the data for $\chi \times H$ are shown as a sequence of curves, each for a different T. The curve for the highest T corresponds to 187.4 K and the lowest to 120 K. The change in the curves as the temperature decreases is obvious. As T approaches the LP, the singularity becomes sharper. The decay of χ as we enter into the P phase also becomes less abrupt as T decreases, in spite of the lower temperature. Both effects reflect the variation of α along the boundary. In Fig. 4 the curves for $T = 170.5$ and 121.4 K are compared.

The experimental data, previously corrected for the demagnetization factor of the sphere, were fitted to the expression

$$\chi H = \chi_0 H + \frac{A}{\alpha}(h^{-\alpha} - 1), \tag{5}$$

where $h = |H - H_\lambda|/H_\lambda$. The regular term $\chi_0 H$ is necessary, since in the absence of the singular contribution, the susceptibility should be constant inside the F phase ($H < H_\lambda$) and

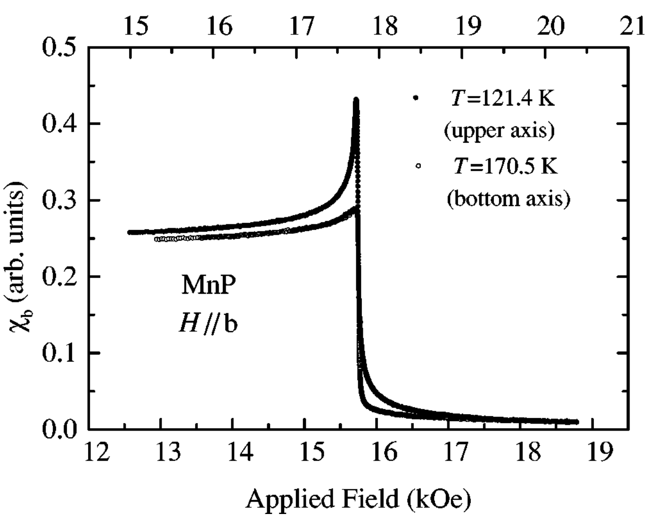

FIG. 4. Comparison of two traces obtained at temperatures well above (170.5 K) and very close to the LP (121.4 K). Note the different horizontal axes.

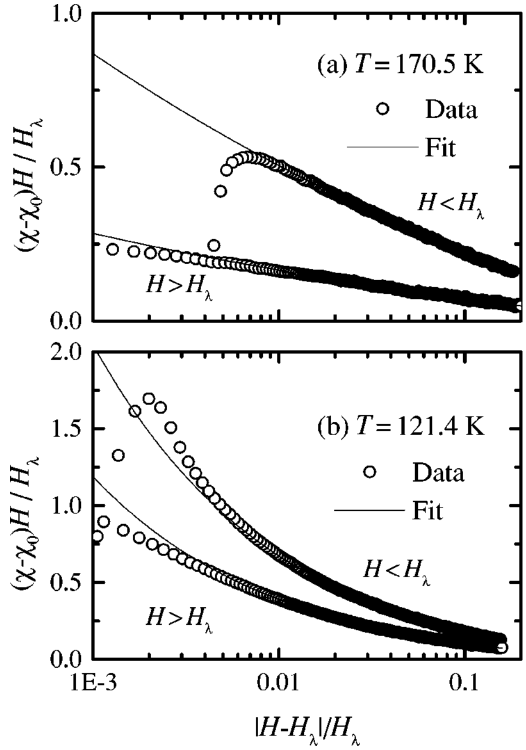

FIG. 5. Plots of the nonlinear fits to Eq. (5) for data with (a) $T = 170.5$ K and (b) $T = 121.4$ K. The corresponding parameters are listed in Table I.

very small in the disordered phase ($H > H_\lambda$). The singular term on the right-hand side contains the essential singularity of the Riedel-Wegner expression [Eq. (2)], in a form widely used in the analysis of specific heat data.[12]

We designate the fit parameters corresponding to data points sets above ($H > H_\lambda$) and below ($H < H_\lambda$) the transitions with superscripts (+) and (−), respectively. Nonlinear least-squares fits were performed to adjust the data and obtain values for $\chi_0^\pm$, $A^\pm$, $\alpha^\pm$, and $H_\lambda^\pm$. As shown in Ref. 8, consistent values for $\alpha^\pm$ and $H_\lambda^\pm$ with unconstrained fits are obtained only close to the LP, where the singularity is more pronounced and the phase boundary is nearly ''parallel'' to the T axis. At higher T the λ line becomes more inclined and the ''true'' thermal axis becomes inclined relative to h. Besides, the rounding effect becomes more pronounced. Due to these problems, we made the option in the analysis to perform only constrained nonlinear least-squares fits with the conditions $\alpha^+ = \alpha^-$ and $H_\lambda^+ = H_\lambda^-$.

In Fig. 5 the fits to the singular part of the susceptibility

TABLE I. Numerical results of the constrained fits for five selected temperatures along the F-P λ line.

T (K)	α	H_λ (kOe)	A^+/A^-	χ_0^+ (a.u.)	χ_0^- (a.u.)	σ_α
121.4	0.443	16.38	0.580	1.86	0.003	0.08
125.6	0.352	16.25	0.511	1.85	0.009	0.09
132.4	0.270	16.05	0.435	1.80	0.018	0.12
154.0	0.117	15.22	0.358	1.75	0.012	0.16
170.5	0.108	14.63	0.327	1.74	0.031	0.18

for 121.4 and 170.5 K are compared with the data. The range of the fits extends from $h \approx 0.1$ to $h \approx 0.01$, being narrower at the higher temperatures due to the rounding of the transition. This strong rounding causes a rapid decrease of the exponent, yielding even a negative value at the highest temperature (187 K) and an appreciable increase in the error of the fit.

In Table I the results are listed for five selected temperatures. The error in α (σ_α in the last column of the table) increases at higher temperatures due to rounding in the transition. However, the tendency of the exponent α to approach the Ising value ($\alpha = 0.125$) as T increases along the λ line is clear. The value obtained at $T = 121.4$ K is in good agreement with the theoretically predicted value for the uniaxial LP. The value of the amplitude ratio near T_L is close to that expected for the Ising model ($A^+/A^- = 0.55$), but seems to be more sensitive to the rounding effects and departs considerably from the expected value at the highest T.

In conclusion, we have shown an evidence for the Lifshitz-Ising crossover along the F-P λ line in a transverse field in MnP.

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*Electronic address: cbecerra@if.usp.br

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