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Specific-heat exponent and critical-amplitude ratio at the Lifshitz multicritical point

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The specific-heat critical exponent α_L associated with the uniaxial Ising-type Lifshitz point (LP) in MnP, was determined experimentally from the study of the magnetic susceptibility χ perpendicular to the easy axis, as a function of the magnetic field. According to generalized scaling theory, the divergence of χ, at the ferromagnetic-paramagnetic phase boundary, near the LP, is governed by α. We found α_L between 0.4 and 0.5, in agreement with estimates from other experimentally known critical exponents in MnP. For the critical-amplitude ratio we found $A^+/A^- = 0.65 \pm 0.05$.

Lifshitz multicritical points (LP's) arise in magnetic systems with competing interactions at the confluence of a modulated and a ferromagnetic phase with the paramagnetic phase. The transitions from the ordered phases to the paramagnetic one are second order. Along the modulated segment of this second-order line (λ line), the wave vector **q** (with m components) varies continuously and vanishes at the LP.[1] Experimentally pure Lifshitz critical behavior has only been reported in the orthorhombic magnetic system MnP,[2,3] which exhibits a uniaxial Ising-type LP ($d=3$, $n=1$, $m=1$). Another type of Lifshitz behavior has been suggested in the structural transition of $RbCaF_3$,[4] which is near a tricritical Lifshitz point (TLP). In this Rapid Communication we report the first experimental determination of the specific-heat exponent (α_L) and its corresponding critical amplitude ratio for a LP. The measurements were carried out in MnP.

Previous studies made in MnP (Refs. 2 and 5) stressed its connection with the $d=3$ anisotropic next-nearest-neighbor Ising model[6] that shows a uniaxial LP in the T vs κ plane (T is temperature and $\kappa = J_2/J_1$ is the ratio of competing antiferromagnetic and ferromagnetic exchange interactions). In mean-field theory $\kappa_L = 0.25$ at the LP. In MnP, the modulated phase is a fanlike one,[2] with the wave vector **q** in the **a** direction (the hard magnetic axis). Neutron-diffraction measurements of spin-wave dispersion curves in MnP,[7] interpreted in terms of a model with interactions J_1 and J_2, respectively, between nearest and next-nearest 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. It was also shown,[2] based on generalized scaling theory,[8] that, near the LP, the thermal scaling axis may be taken as a magnetic field H perpendicular to the easy axis (c axis). This leads to the prediction that the susceptibility $\chi = \partial M/\partial H$, measured as a function of H at constant T, diverges at the transition as $\chi \propto |H-H_c|^{-\alpha}$, where α is the specific-heat critical exponent. We use this result to investigate α_L, by means of susceptibility measurements.

Theoretical predictions give large values for α_L. Renormalization-group (RG) calculations, to first order in $\epsilon_L = 4+m/2-d$, predict[1] $\alpha_L = 0.25$ for $d=3$, $n=m=1$. A larger value is expected in MnP in view of the following analysis. In the case of a LP, the spin-spin correlations are described by two pairs of critical exponents, (η_{L4}, ν_{L4}) and (η_{L2}, ν_{L2}), which are related through $(4-\eta_{L4})\nu_{L4} = (2-\eta_{L2})\nu_{L2}$ and the hyperscaling relation $2-\alpha_L = m\nu_{L4} + (d-m)\nu_{L2}$. From scaling[9] $\nu_{L4} = \phi\beta_k$. The crossover exponent ϕ was determined experimentally in MnP,[2] as $\phi = 0.63 \pm 0.04$, in close agreement with the first-order RG prediction[9] $\phi = 0.625$. The exponent β_k is associated with the vanishing of **q** along the modulated segment of the λ line as the LP is approached. Neutron-diffraction studies in MnP,[3,10] along the fan-paramagnetic transition line between 4 and 118 K, resulted in $\beta_k = 0.480 \pm 0.013$, in good agreement with the value $\beta_k = 0.5$ predicted from first-order RG (Ref. 1) and series expansions.[11] The above experimental values lead to $\nu_{L4} = 0.30 \pm 0.02$, which also agrees with the first order in ϵ_L estimate,[1] $\nu_{L4} = \nu_{L2}/2 = 0.3125$. With $\eta_{L4} = \eta_{L2} = 0$ (true up to first order in ϵ_L), $\alpha_L = 0.49 \pm 0.10$. To second order in ϵ_L,[9] $\eta_{L2} = -2\eta_{L4} = \frac{1}{12}$, and $\alpha_L = 0.42 \pm 0.11$. Very large values for α (larger than 0.5) are also predicted by RG at the TLP.[12]

Measurements of the ac susceptibility (χ' and χ'') were performed on a MnP single crystal, cut into a 5.2-mm-diam sphere. Both the applied field H_0, provided by a superconducting magnet, and the modulation field (2.5 Oe amplitude and 17 Hz) were parallel to the b axis. A very careful alignment of H_0 was made, using the shape of the susceptibility curves at the ferromagnetic-paramagnetic transition near the LP. The final alignment was estimated to be better than 0.05° in the bc plane and 0.5° in the ab plane. The runs were made at fixed T, within ± 0.01 K, and with H_0 varying at 3.4 Oe/s. In Fig. 1 we show some representative χ' vs H_0 curves, and the obtained phase boundaries near the LP. In curve a, taken below T_L, the large spike corresponds to the first-order ferromagnetic-fan transition. The other peak marks the second-order transition to the paramagnetic phase. As T_L is approached, these transitions get closer in field, until they collapse in a single λ-shaped anomaly, characteristic of a strong divergence in χ' (curve b). At higher T this anomaly gets less and less pronounced and rounding effects be-

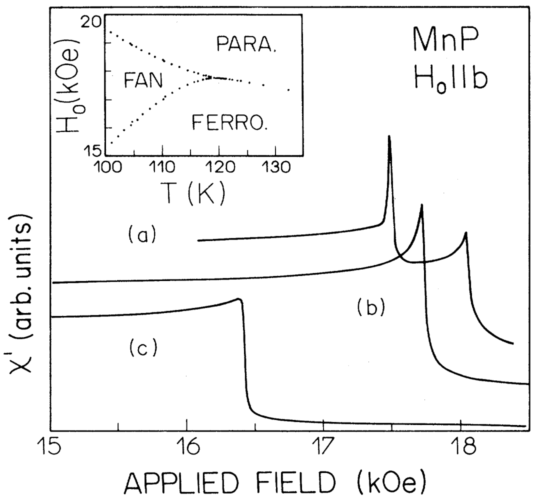

FIG. 1. Traces of the in-phase ac susceptibility vs applied magnetic field at various temperatures: Curve (*a*), $T=114.6$ K; curve (*b*), $T=121.4$ K; curve (*c*), $T=154$ K. The curves are displaced vertically for more clarity. Inset: Phase diagram near T_L, determined from the susceptibility measurements.

come evident, as illustrated by curve *c*.

To locate T_L we used the χ'' vs H_0 data. At the ferromagnetic-fan transition, this susceptibility exhibits a narrow and well defined peak. This peak is due to the loss mechanisms present in a first-order transition and vanishes as the first-order line goes into a second-order one, at T_L. Figure 2 shows the height of this peak, $\Delta\chi''$, as a function of T. The abrupt decrease can be readily extrapolated to zero, yielding $T_L=(120.8\pm0.3)$ K. This value is in excellent agreement with all previous determinations, from fits of the phase boundaries near the LP,[2] which can be summarized as $T_L=(121\pm1)$ K.

We used previously reported magnetization data[13] to obtain the internal susceptibility χ and field H from χ' and H_0. The experimental data were fitted to the expression:

$$C^* \equiv \chi H = \chi_0 H + \frac{A}{\alpha}(|h|^{-\alpha}-1),$$

with $h=|H-H_c|/H_c$. Superscripts (+) and (−) will denote the adjusted parameters for $H>H_c$ and $H<H_c$, respectively. The second term on the right-hand side of this expression is widely used in the analyses of specific-heat data.[14] The regular term $\chi_0 H$ is necessary, since in the absence of the singular contribution, χ should be a constant inside the ferromagnetic phase ($H<H_c$) and very small inside the disordered phase ($H>H_c$).[5] We employed nonlinear least-squares fits to adjust $\chi_0^{\pm}$, $A^{\pm}$, $\alpha^{\pm}$, and $H_c^{\pm}$ independently, and also with the constraints $H_c^+=H_c^-$ and $\alpha^+=\alpha^-$. A study of the dependence of the adjusted parameters on the fit range (defined between h_{min} and h_{max}) was performed for all the curves analyzed. The error bars quoted in all figures are estimates based on these analyses, whose details will be published elsewhere.

Figure 3 illustrates the results of the independent fits in the case of a curve very near T_L. Figure 3(a) shows the experimental points as a plot of $C_S^*=(\chi-\chi_0)H$, the singular part of C^*, as a function of $\log_{10}(h)$. In Figs. 3(b) and 3(c) we see the deviations from the best fit for the (−) and (+) branches of the data. The adjusted parameters are given in the caption. For all the curves near T_L, the best values of H_c^- and H_c^+ agree within 5 Oe ($\approx 3\times10^{-4}H_c$). The intervals of the fits ranged, typically from $h_{min}\approx 4\times10^{-3}$ to $h_{max}\approx 2\times10^{-1}$. At higher T the range is reduced, as the rounding effect becomes more pronounced. The fits, however, always extended to more than 1.5 decades in the reduced field h. The rms deviation

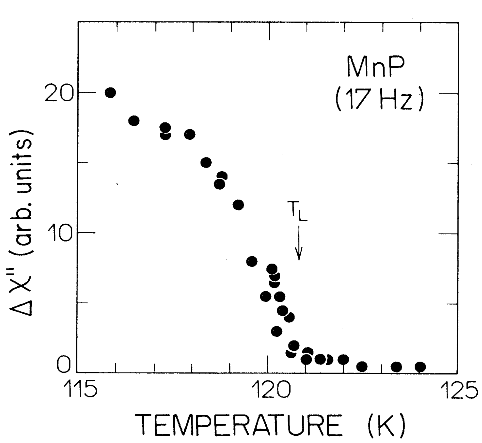

FIG. 2. Height of the peak in χ'' at the transitions from the ferromagnetic phase as a function of T for the determination of T_L.

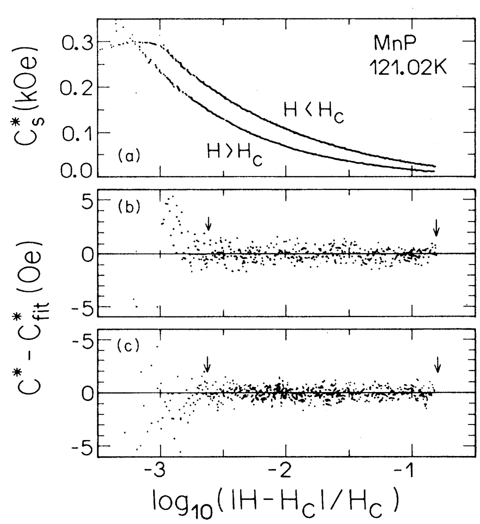

FIG. 3. (a) Singular part of χH vs reduced internal field; (b) and (c) deviations from the best fit, respectively, for $H<H_c$ and $H>H_c$. The adjusted parameters and their standard errors are (b) $H_c^-=16.380\pm0.001$ kOe, $\chi_0^-=(1.859\pm0.001)\times10^{-2}$ (cgs), $A^-=8.76\pm0.03$ Oe, $\alpha^-=0.371\pm0.002$; (c) $H_c^+=16.380\pm0.001$ kOe, $\chi_0^+=(8\pm1)\times10^{-5}$ (cgs), $A^+=3.80\pm0.01$ Oe, $\alpha^+=0.509\pm0.005$. The arrows indicate the limits of the fits.

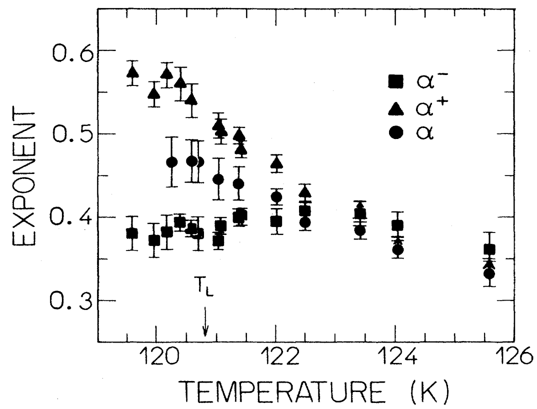

FIG. 4. Values obtained for the critical exponent near T_L. α^- and α^+ resulted from the independent fits of the two branches of data. α is from the fits with the constraints $\alpha^+=\alpha^-$ and $H_c^+=H_c^-$.

for all these fits are about 5×10^{-4} kOe, less than 1% of the representative values of C_S^*.

In Fig. 4 we show α^+ (triangles) and α^- (squares) as a function of temperature. Above 122.5 K these two exponents agree within 5% and decrease with increasing T. Below this temperature, however, substantial differences between them are observed: α^- reaches 0.41 around 122 K, and then decreases slowly, while α^+ continues to increase, reaching 0.55 around 120 K. At $T_L=120.8\pm0.3$ K, we found $\alpha^-=0.39\pm0.02$ and $\alpha^+=0.53\pm0.03$.

At the lowest temperatures analyzed, the presence of the first-order ferromagnetic-fan transition is evident in the data for $H<H_c$. It is seen in the analysis as a sudden rise of the deviations, towards the positive side of the best fit, which restricts $h_{\min}$. Vestiges of these effects persist in the curves up to 122 K [see Fig. 3(b)]. Above 122 K, $h_{\min}$ is limited by rounding effects, which cause gradual and negative systematic deviations from the best fit. The fact that the curves in which the vestiges of the first-order transition are present, are the same for which α^+ results significantly different from α^-, suggests that the analytic behavior of χ is affected by the closeness of the ferromagnetic-fan transition. This may be the cause of the observed discrepancies for α.

In Fig. 4 we also show the values obtained for α (circles) from the fits with the constraints $\alpha^+=\alpha^-$, required by scaling theory, and $H_c^+=H_c^-$. The quality of these fits, however, is not as good as the previous ones. The corresponding deviation plots show systematic departures from the adjusted curves. These departures are always in the 1–2% range of C_S^* and are seen as a slight curvature in the strip of fitted data, downward for the $(-)$ branch and upward for the $(+)$ branch. This curvature disappears only above 122 K, where the previous unconstrained fits already yielded the same α for both sides of the transition.

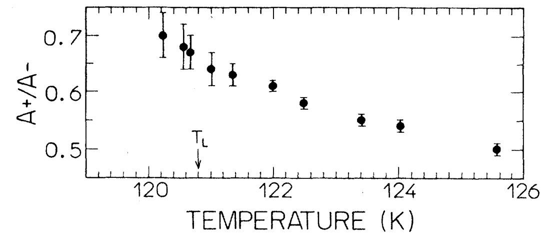

FIG. 5. The critical amplitude ratio A^+/A^-, from the constrained fits, as a function of T.

From the analyses made, it can be established that, at the LP, the specific-heat exponent is quite high, in the range of 0.4–0.5. The different values for the exponents yielded by the unconstrained fits, near T_L, may represent a departure from theoretical predictions, but also may be attributed to the closeness of the first-order ferromagnetic-fan transition. The fits with the $\alpha^+=\alpha^-$ condition resulted in an intermediate value, namely $\alpha_L=0.46\pm0.03$.

For $T>T_L$, along the ferromagnetic-paramagnetic λ line, a crossover to an Ising-type critical behavior, for which α should be near 0.125, is expected. As can be seen in curve c of Fig. 1, the divergence in χ' is sensitively weaker at high temperatures. In fact, the measured exponents decrease with increasing temperature (above T_L), and analyses of curves above 150 K yielded values for α in the 0.1–0.2 range.

From the constrained fits, we obtained also the critical amplitude ratio A^+/A^-, plotted in Fig. 5 as a function of T. We obtained, at T_L, $A^+/A^-=0.65\pm0.05$. No theoretical estimate has been made for this universal ratio in the case of Lifshitz points. Our result is comparable to calculated[15] and observed[16] values for the Ising-model critical point.

In conclusion, we have determined for the first time, from the divergence in χ, the specific-heat exponent and the critical amplitude ratio near a uniaxial LP. Our values for α_L are higher than the first order in ϵ_L result ($\alpha_L=0.25$), but agree with the estimates based in experimental values for ϕ and β_k in MnP.

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