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ABSTRACT

We have measured the variation of $\vec{q}$ in the fan phase of MnP in order to test whether the para-ferro-fan triple point is a Lifshitz point. Along the para-fan phase boundary, $\vec{q}$ continuously decreases as the triple point is approached, extrapolating to zero at a temperature in good agreement with other measurements of the triple point. The temperature dependence of $\vec{q}$ along this phase boundary is in approximate agreement with theoretical expectations for Lifshitz point behavior. These data support the conclusion that the triple point is a Lifshitz point.

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INTRODUCTION

Many theoretical studies of the Lifshitz point (LP) have been published in recent years (1), but there has been a notable absence of positive experimental evidence on the existence of this effect. In a magnetic system, this multicritical point has the following three essential characteristics. (i) The LP is a meeting point of three phases: a paramagnetic phase, a ferromagnetic phase (2), and a helicoidal phase with a variable equilibrium wave vector which is a function of thermodynamic parameters. (ii) The transitions from the paramagnetic phase to both ordered phases should be of second order. (iii) The wave vector $\vec{q}$ in the helicoidal phase should approach zero continuously as the LP is approached.

Based on a study of the phase diagram and on susceptibility measurements, it was recently concluded that the para-ferro-fan triple point of MnP is a LP (3). However, neither the phase diagram nor the susceptibility give direct evidence on the variation of $\vec{q}$ in the fan phase. The purpose of the present work was to measure the variation of $\vec{q}$ in the fan phase, and to check whether indeed $\vec{q} \rightarrow 0$ as the LP is approached. We also attempted to determine the critical exponent

β_k which governs the variation of $\vec{q}$ along the para-fan phase boundary.

MnP has an orthorhombic cell and is ferromagnetic below 291 K, with a magnetic phase change at 50 K (4). Neutron diffraction studies (5,6) have shown the low temperature (zero field) structure to be a spiral with the wave vector along $\vec{a}$ (a > b > c). Application of a magnetic field along $\vec{b}$ produces a fan phase, as shown by magnetization (7) and neutron diffraction studies (8). The Mn moments are ferromagnetically coupled in planes perpendicular to $\vec{a}$, but the direction of these moments wobbles about the $\vec{b}$ direction in a periodic fashion with a wave vector

$$\vec{q} = 2\pi \frac{\delta}{a} \hat{a}. \tag{1}$$

We have measured the position and intensity of the resulting magnetic satellites at the $(2 \pm \delta\ 0\ 0)$ positions as functions of applied field and temperature (89 to 117 K).

EXPERIMENTAL DETAILS

The experiments were performed at the High Flux Isotope Reactor, using the HB-1 spectrometer in the unpolarized beam, triple-axis mode. The single-crystal sample (2.6 x 2.6 x 0.9 mm with the long dimension along b and c) was mounted in the gap of a superconducting solenoid with the b axis parallel to the vertical magnetic field. The (002) reflection from pyrolytic graphite was used at the monochromator and analyzer positions with 40' collimation before and after the sample. With an incident neutron wavelength of 2.016 Å, this gave adequate resolution (peak widths of 0.038/a) for temperatures below 114.6 K. For the 117 K data the collimation was reduced to 20', the wavelength increased to 2.443 Å, and the (004) reflection was used at the analyzer position. This resulted in peak widths of 0.023/a. Observations at 114.6 K were made with both resolution conditions with excellent agreement on the value of δ.

The basic experiment consisted of a longitudinal, elastic scan through the (200) position. At a given temperature the phase diagram of Ref. 3 was consulted to select a starting field in the ferromagnetic phase, slightly below the ferro-fan boundary. Keeping the temperature fixed, the scan was repeated as the field was increased in steps until magnetic satellites were no longer visible.

A favorable case is illustrated in Fig. 1 which shows the data for 89.1 K and 13 kOe. Note that the second-order satellites at $(2 \pm 2\delta\ 0\ 0)$ are easily observed in this case. It is very difficult to observe these satellites at temperatures above 100 K or at fields near the fan-para phase boundary. Values of δ were obtained from plots similar to Fig. 1 by measuring the change in ξ between the half-peak points on the outer sides of the first-order satellites. This gives

$$\Delta\xi = 2\delta + w, \tag{2}$$

where w is the peak width. As the temperature is increased the satellites move toward the central (200) peak and the satellite intensity decreases, resulting in a difficult experimental situation. Difference plots between $I(H,T)$ and $I(H_F,T)$, where H_F is in the ferromagnetic region, were useful when the satellite peaks were very weak or very close to the (200). There may be a slight tendency for broadening of the magnetic satellites as the paramagnetic phase is entered.

RESULTS

Our results are summarized in Fig. 2 which shows the satellite peak intensity and δ as functions of field and temperature. The intensity curves may be used to construct the phase diagram. At a fixed temperature and low applied field, the system is ferromagnetic and the satellite intensity is zero. The abrupt increase marks the ferro-fan phase boundary at H_1. In the fan phase the intensity rises rapidly with increasing field and then gradually falls to zero at the fan-para boundary. There is some ambiguity in the location of this boundary, as indicated by the 89.1 K data, because the intensity gradually tails off in the paramagnetic phase, indicating critical behavior. We have used an intuitive extrapolation, as indicated in Fig. 2, to locate this phase boundary at H_2. Our results for $H_1(T)$ are in excellent agreement with magnetostriction results on the same sample, but $H_2(T)$ as determined from Fig. 2 is consistently higher, by roughly 600 Oe, than the corresponding magnetostriction result. We believe that our extrapolation of the neutron intensities may lead to H_2 values which are slightly high, but there may be also a real difference attributable to the completely different experimental techniques.

The linear behavior of δ with applied field at fixed temperature is evident in Fig. 2 as is the dramatic decrease in δ as the temperature is increased. We attempted an observation at 118 K but the combination of low satellite intensity and small δ produced unreliable results--that is, the satellites were lost in the tails of the (200) peak.

DISCUSSION

The data of Fig. 2 suggest two independent measures of the critical temperature above which the fan phase does not exist. One is the vanishing point of the magnetic width ($H_2 - H_1$) of the fan phase and the other is the vanishing point of the maximum satellite intensity observed at a fixed temperature, $I_M(T)$. The maximum amplitude of the oscillating c-axis component of the Mn moments is proportional to $I_M^{\frac{1}{2}}$. The temperature dependence of $I_M^{\frac{1}{2}}$ and of the magnetic width of the fan phase are shown in Fig. 3. Both curves yield reasonable extrapolations in excellent agreement with the LP of Ref. 3, $T_L = 121 \pm 1$ K.

The theory of the LP predicts a characteristic variation of $\vec{q}$ along the para-helicoidal segment of the λ line. In the theoretical papers the phase diagram is in the P-T plane, where P is a general thermodynamic parameter. Very close to the LP and along the para-helicoidal boundary, $\vec{q}$ is predicted to vary as $(P - P_L)^{\beta_k}$, where P_L is the value of P at the LP. In mean-field theory $\beta_k = 0.5$, and to a better approximation $\beta_k = 0.54$ (1,9). To translate this theoretical prediction to the present case, where the phase diagram is in the T-H plane, it is necessary to introduce scaling axes as discussed in Ref. 3. Using their procedure we expect that near the LP on the fan-para boundary,

$$\delta = K(T_L - T)^{\beta k}, \qquad (3)$$

where T_L is the temperature at the LP.

Using the data of Fig. 2 we have determined $\delta(T)$ along the fan-para boundary and have made a least-squares fit of Eq. 3 with K, T_L and β_k as adjustable parameters. The results are displayed in Fig. 4 with $\beta_k = 0.44 \pm 0.05$ and $T_L = 121.7 \pm 1.3$ K. Recalling the difficulty in exactly determining the fan-para boundary, we have also used the phase diagram of Ref. 3 and our measurements of δ to establish $\delta(T)$ along the λ line. This results in a curve very similar to Fig. 4 with $\beta_k = 0.49 \pm 0.03$ and $T_L = 122.6 \pm 0.9$ K. It is not clear that the theoretical power law should extend over our temperature range, so that our value of β_k may only be approximate. Nevertheless, it appears that the temperature dependence of $\vec{q}$ is in approximate agreement with the theoretical prediction for LP behavior.

SUMMARY

We have measured the variation of $\vec{q}$ in the fan phase of MnP as a function of magnetic field and temperature between 89 K and 117 K. Over this temperature range the wavelength of the magnetic periodicity was found to vary from 12.6 lattice units along $\vec{a}$ (75 Å) to 32.2 lattice units (191 Å). Along the para-fan boundary, $\vec{q}$ decreases as the triple-point is approached, extrapolating to zero at a temperature close to the triple point of Ref. 3. The temperature dependence of $\vec{q}$ along this phase boundary is in approximate agreement with the theoretical expectation for a LP. Thus, our data are consistent with the interpretation that the para-ferro-fan triple point in MnP is a Lifshitz point.

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b. Supported by the National Science Foundation.
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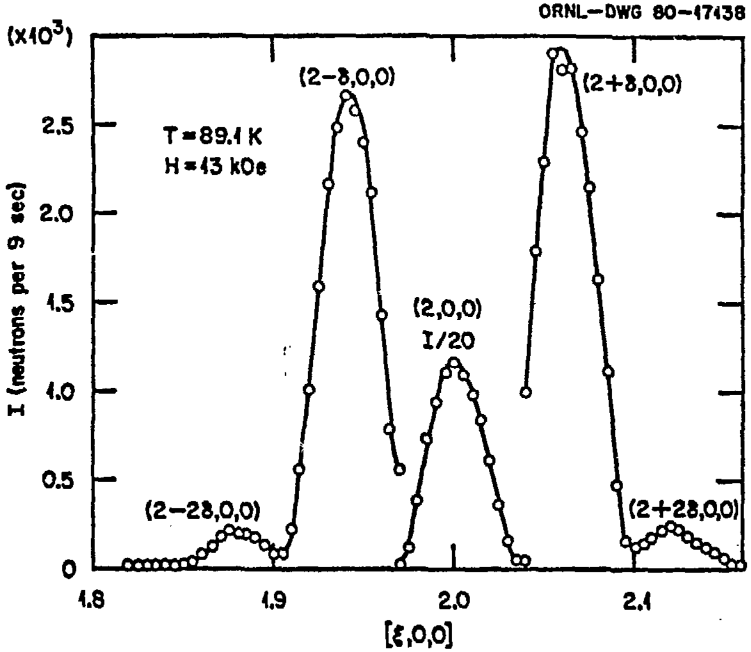

Fig. 1. An elastic triple-axis scan through the (200) position of MnP showing first and second-order magnetic satellites. Note that the central (200) peak is shown on a reduced scale.

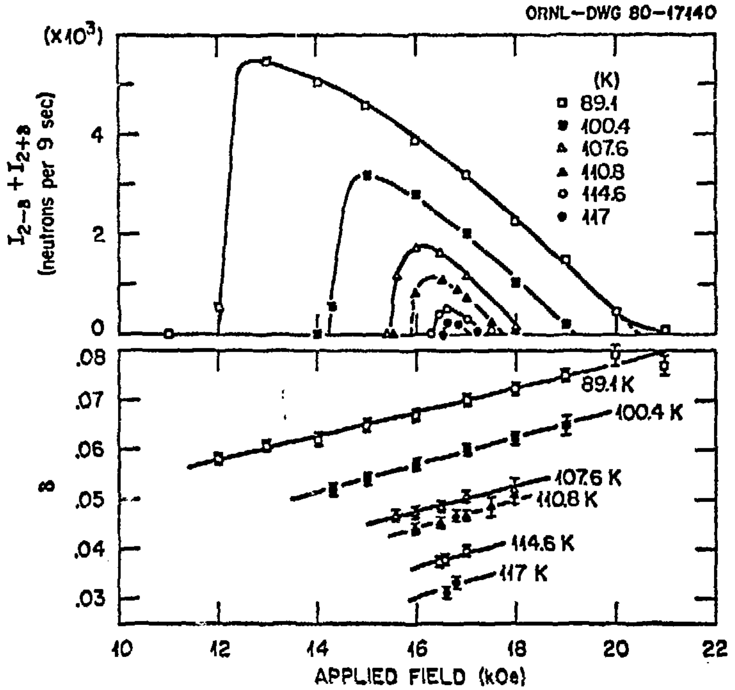

Fig. 2. Satellite peak intensity and wave vector as functions of applied field and temperature.

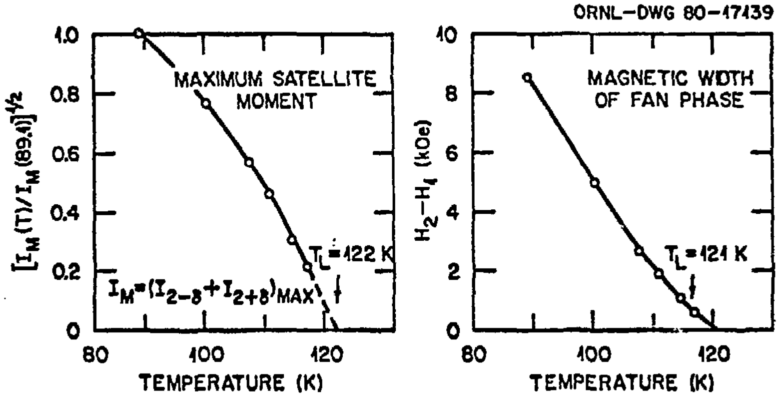

Fig. 3. The temperature dependences of the normalized maximum c-axis moment and of the magnetic width of the fan phase give triple points in agreement with Ref. 3.

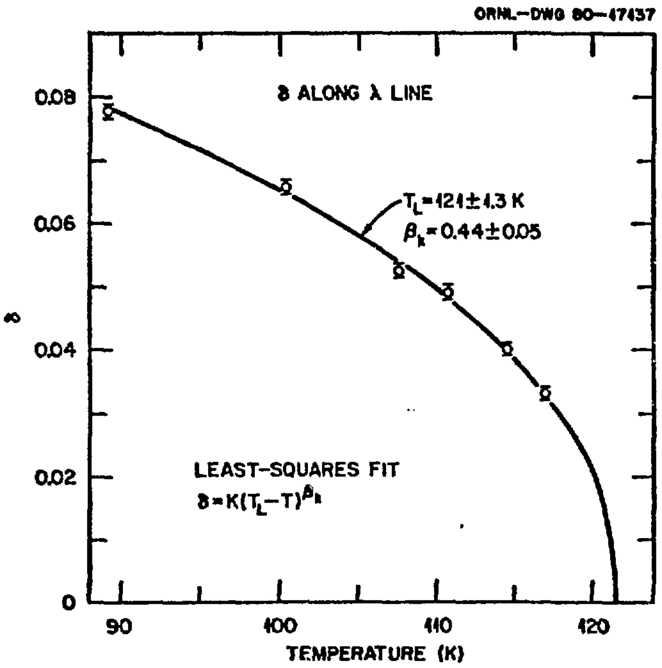

Fig. 4. Temperature dependence of δ along the fan-para phase boundary. The solid line is a least-squares fit of the theoretical relation to the experimental data.