Theoretical interpretation for the occurrence of a Lifshitz point in MnP

C. S. O. Yokoi
Instituto de Física, Universidade de São Paulo, C.P. 20.516, São Paulo, Brazil

M. D. Coutinho-Filho
Departamento de Física, Universidade Federal de Pernambuco, 50.000 Recife, Brazil

S. R. Salinas
Instituto de Física, Universidade de São Paulo, C.P. 20.516, São Paulo, Brazil
(Received 11 June 1981)

We present a theoretical interpretation for the occurrence of a Lifshitz point in the field-temperature phase diagram of MnP. Our mean-field calculations, which are based on a localized spin Hamiltonian, are in qualitative agreement with recently reported experiments. In particular, we obtain asymptotic expressions for the phase boundaries, which meet tangentially at the Lifshitz point, and for some other thermodynamic quantities of interest, such as the longitudinal and transverse susceptibilities. A renormalization-group argument also supports the conclusion that the Lifshitz point in MnP is of a uniaxial one-component type.

In a recent publication Becerra *et al.*[1] have reported measurements of the field-temperature phase diagram of MnP in the neighborhood of the "triple point" separating the paramagnetic, the ferromagnetic, and the fan phases. Based on measurements of the transverse differential susceptibility they suggested that the "triple point" is a Lifshitz point[2] and estimated the scaling crossover exponent $\phi = 0.634 \pm 0.03$ from the shape of the phase boundaries which meet tangentially at the Lifshitz point. This value of ϕ as well as the shape of the phase boundaries are expected on theoretical grounds[3,4] for a Lifshitz point characterized by the lattice dimensionality $d = 3$, a one-component order parameter $(n = 1)$, and a unique direction of the wave-vector instability $(m = 1)$.

Despite the fact that the Lifshitz point in MnP seems to belong to the same unversality class of simple model systems such as the axial next-nearest-neighbor Ising model[5] (or ANNNI model), it nevertheless presents some remarkable particularities. For instance, since Lifshitz points are usually associated with Tp phase diagrams, where T is temperature and p is related to pressure or material composition, it is not straightforward to conclude that a Lifshitz point should be found in field-temperature phase diagrams.[6] In this respect we mention that the ANNNI model does not exhibit a Lifshitz point in the field-temperature phase diagram.[7] It is also noteworthy that the modulated phase in MnP is a fan phase, which has never been considered in the previous theoretical studies of Lifshitz points. These facts then suggest that a proper understanding of the multicritical behavior of MnP near the "triple point" requires the consideration of a particular Hamiltonian which is suitable for this magnetic crystal.

In this Communication we report a possible interpretation for the experimental results of Becerra *et al.* on the basis of a localized spin model for the thermodynamic behavior of MnP. Although some properties of this compound are better accounted for by a band model of itinerant spins,[8] we remark that a localized spin model has been successfully applied to explain many magnetic properties of MnP.[9] The statistic-mechanical calculations in the mean-field approximation, together with a renormalization-group analysis, give results in qualitative agreement with the experimental measurements of Becerra *et al.* and do support their suggestion concerning the nature of the "triple point."

MnP is a magnetic compound with orthorhombic structure $(a > b > c)$, in which the c axis is easy, the b axis intermediate, and the a axis extremely hard. In the absence of an applied field, MnP is ferromagnetic between 47 and 291 K, with moments parallel to the easy axis. Below 47 K a screw phase is observed in which the moments rotate in the bc plane with propagation vector $\vec{q}$ along the a axis. Another phase called fan is observed when a magnetic field $\vec{H}$ is applied along the b axis, as in the experiments of Becerra *et al.* In this fan phase the moments are still in the bc plane but do not undergo a full rotation.[10] The field-temperature phase diagram of MnP in the neighborhood of the "triple point" is drawn schmatically in Fig. 1. Experiments indicate that the ferro-fan transition is discontinuous, while the para-ferro and the para-fan transitions are of second order.[1,10]

Hiyamizu and Nagamiya[9] have interpreted the magnetization process in MnP on the basis of a model which essentially takes into account the com-

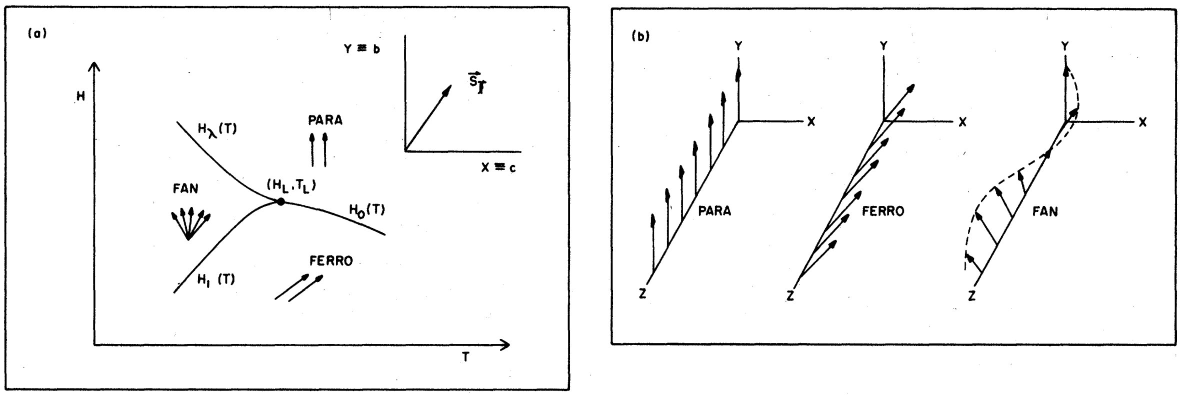

FIG. 1. (a) Schematic representation of the field-temperature phase diagram of MnP in the neighborhood of the "triple point." The phase boundaries meet tangentially at (H_L, T_L). $H_\lambda(T)$ and $H_0(T)$ are lines of critical points. $H_1(T)$ is a line of first-order transitions. The magnetic field is applied along the $b \equiv y$ direction, and the moments are in the bc plane. The propagation vector in the fan phase is along the $a \equiv z$ direction. (b) Representation of the average magnetization per spin in successive layers, in the ferromagnetic, paramagnetic, and fan phases. In the fan phase, it should be noticed that the x component of the magnetization exhibits oscillating behavior along the z direction.

peting exchange interactions along the a axis and the anisotropy in the bc plane. We will basically assume the same model, but since our aim is directed towards the understanding of the phenomenon rather than to the quantitative analysis of experimental results, we will disregard those features of the system which are not essential to the present work. Accordingly, we write the following XY Hamiltonian,

$$H = -\tfrac{1}{2} \sum_{\vec{l}, \vec{l}'} J(\vec{l} - \vec{l}')\vec{S}_{\vec{l}} \cdot \vec{S}_{\vec{l}'} - D \sum_{\vec{l}} [(S^x_{\vec{l}})^2 - (S^y_{\vec{l}})^2] - H \sum_{\vec{l}} S^y_{\vec{l}} \ , \qquad (1)$$

where $\vec{l} \equiv (l,m,n)$ is the lattice vector of a simple cubic lattice with N points in each direction, $\vec{S}_{\vec{l}} = (S^x_{\vec{l}}, S^y_{\vec{l}})$ are Pauli spin-$\frac{1}{2}$ operators, and the anisotropy factor $D > 0$ favors the x direction (c axis). The exchange constants $J(\vec{l} - \vec{l}')$ are ferromagnetic in the xy (cb) plane and include the effects of competing interactions along the z direction (a axis). We remark that the choice of spin-$\frac{1}{2}$ operators is deliberate, since the localized spin moments of MnP are not well defined and also because the magnitude of the spin is believed to be inessential to the critical behavior. We also observe that this XY Hamiltonian had already been subjected to a theoretical investigation by Kitano and Nagamiya[11] two decades ago, but of course they had not focused their attention on the problem of the Lifshitz point.

In the mean-field approximation, in which one effective field is assigned to each layer n, the Gibbs free-energy may be written as

$$N^{-3}G(T,H,N;\{\vec{M}_n\}) = -kT\ln 2 + \frac{kT}{2N}\sum_n [(1+M_n)\ln(1+M_n) + (1-M_n)\ln(1-M_n)] - \frac{1}{2N}\sum_{n,n'} J(n-n')\vec{M}_n \cdot \vec{M}_{n'} - \frac{D}{N}\sum_n [(M^x_n)^2 - (M^y_n)^2] - H\sum_n M^y_n \ , \qquad (2)$$

where the average magnetization per spin in layer n, $\vec{M}_n = (M^x_n, M^y_n)$, is given by the solution of the system of coupled equations

$$\tfrac{1}{2}kT\frac{M^\alpha_n}{M_n}\ln\left(\frac{1+M_n}{1-M_n}\right) = \sum_{n'} J(n-n')M^\alpha_{n'} \pm 2DM^\alpha_n + H^\alpha_n \ . \qquad (3)$$

The plus and minus signs correspond to $\alpha = x$ and $\alpha = y$, respectively, with $H^x_n = 0$ and $H^y_n = H$. The effective exchange constants $J(n-n')$, which represent the interactions between spins in different layers, are defined by

$$J(n-n') = N^{-2}\sum_{l,m}\sum_{l',m'} J(l-l', m-m', n-n') \ .$$

Since the transition from the paramagnetic phase, with uniform magnetization $\vec{M}_n = (0,M)$, to the ordered phases, either ferromagnetic or fan, is characterized by the onset of a nonzero x component of the magnetization, it is of interest to examine the wave-vector-dependent transverse susceptibility χ^x_q. By considering a perturbation field $H^x_n = \delta h^x_n$ in Eq. (3), we obtain in the paramagnetic region

$$\chi_q^x = [\hat{J}(0) - \hat{J}(q) - 4D + M^{-1}H] \quad , \tag{4}$$

where M is determined by the equation

$$M = \tanh\left(\frac{[\hat{J}(0) - 2D]M + H}{kT}\right) \quad , \tag{5}$$

and $\hat{J}(q) = \sum_n J(n)\exp(iqn)$ is the Fourier transform of the exchange constants. The transition to the ordered phases should be characterized by the divergence of χ_q^x, namely, by the condition

$$\hat{J}(0) - 4D + M^{-1}H = \max_q \hat{J}(q_c) \quad . \tag{6}$$

From this last equation, and on the assumption that $\hat{J}(q)$ is independent of either T or H, it follows that only one ordered phase, ferromagnetic ($q_c = 0$) or fan ($q_c \neq 0$), should be present in the HT phase diagram near the border of the paramagnetic phase. However, it is an experimental fact that both phases are present. Therefore, if the description in terms of Hamiltonian (1) is to be taken for granted, there is no alternative but to accept the dependence of $\hat{J}(q)$ with T or H. Although many experiments do support this kind of dependence, the underlying mechanism which is responsible for this effect is not as yet completely understood.[12] In any event, Eq. (1) should be regarded as an effective Hamiltonian in which these effects have already been taken into account. $\hat{J}(q)$ may thus be expanded about $q = 0$ in the form

$$\hat{J}(q) = \hat{J}(0) - \alpha q^2 - \tfrac{1}{2}\beta q^4 - \cdots \quad , \tag{7}$$

where, due to the occurrence of the Lifshitz point (T_L, H_L) for $\alpha = 0$, we write in leading order $\alpha = \alpha_T(T - T_L) + \alpha_H(H - H_L)$. Also, $\hat{J}(0)$ may be written as $\hat{J}(0) = J_0 + J_T(T - T_L) + J_H(H - H_L)$. We will further assume that β is a constant positive parameter. These assumptions are sufficient for the calculations of the thermodynamic properties of MnP asymptotically close to the Lifshitz point.

From expressions (6) and (7), it is possible to show that the para-ferro transition line $H_0(T)$, in which $q_c = 0$, has the asymptotic form

$$H_0(T) = H_L - A\,\Delta T - B\,\Delta T^2 \quad , \tag{8}$$

where $\Delta T = T - T_L$, whereas the para-fan transition line $H_\lambda(T)$, in which $q_c \neq 0$, is given by

$$H_\lambda(T) = H_0(T) + C\,\Delta T^2 \quad . \tag{9}$$

The prefactors A, B, and C depend on the structural parameters of the effective spin Hamiltonian. From Eq. (9), it is apparent that both critical lines meet tangentially at the Lifshitz point, and that the mean-field scaling crossover exponent is $\phi = \frac{1}{2}$. We may also conclude, from expressions (7) and (9), that on the para-fan transition line the wave vector q_c behaves asymptotically as $q_c^2 \propto (T - T_L) \propto (H - H_L)$. This result, which implies the mean-field value $\frac{1}{2}$ for the exponent β_k, is in fairly close agreement with very recent neutron diffraction data.[13] The ferro-fan first-order transition line, which is given by

$$H_1(T) = H_0(T) - C(\sqrt{6} + 2)\,\Delta T^2 \quad , \tag{10}$$

may be derived by comparing the asymptotic expressions for the Landau expansion of the Gibbs free energy in these two phases. Therefore, the three transition lines meet tangentially at the Lifshitz point, which is in agreement with the experimental data of Becerra *et al.* It is interesting to observe that the ratio $[H_0(T) - H_1(T)]/[H_\lambda(T) - H_0(T)] = \sqrt{6} + 2 \simeq 4.4$, which was first noticed by Michelson[14] in a similar but different context, still holds in the present model. This fact is not so evident as it might seem at first sight since the asymptotic form of the Landau expansion of the free-energy is affected by the higher harmonic components of the magnetization.[7] The experimental result for this ratio, however, falls about 30% short of the theoretical prediction. We tend to attribute this discrepancy to the well-known limitations of the mean-field approximation.

The uniform transverse susceptibility, defined by $\chi^x = \chi^x_{q\to 0}$, obeys the usual Curie-Weiss law across the para-ferro transition line $H_0(T)$; that is, $\chi^x \propto |H - H_0(T)|^{-1}$ for $H \to H_0(T)$. However, it is continuous and shows a finite cusp across the para-fan transition line $H_\lambda(T)$. Incidentally, as we move along the para-fan line, χ^x diverges as $|\Delta T|^{-2}$ for $T \to T_L$. All these features are in agreement with the experiments of Becerra *et al.*

The longitudinal susceptibility, $\chi^y = \partial M^y/\partial H^y$, is given by the constant $1/4D$ throughout the ferromagnetic phase, and shows a discontinuous behavior across the transition lines. Again, these features are in agreement with the experimental results.[1,15] Moreover, this indicates the correctness of the assumption that the anisotropy factor D is practically temperature independent.

We also suggest some expressions that could hopefully be subjected to experimental verification. For instance, the discontinuity of the longitudinal magnetization across the ferro-fan transition line should behave asymptotically as

$$M^y_{\text{ferro}} - M^y_{\text{fan}} \propto (T - T_L)^2 \propto (H - H_L)^2 \quad , \tag{11}$$

while the ratio $H(T)/M^y(T)$ should be equal to the constant $4D$ along the para-ferro critical line, and tend asymptotically to $4D$ as $(T - T_L)^2$ along the para-fan critical line. The behavior of the wave vector as we penetrate into the fan phase is also of interest. From the condition that the free-energy be minimum it follows at constant temperature the asymptotic expression $q - q_c \propto H - H_\lambda(T)$, where q_c is the critical wave vector on the para-fan critical line.

A renormalization-group analysis of Hamiltonian (1) may be carried out alongside the same techniques devised by Nelson and Fisher[16] to deal with metamagnetism. It is then possible to show that after a few iterations of the renormalization group the Hamiltonian assumes essentially the form of the uniaxial ($m = 1$) one-component ($n = 1$) Landau-Ginsburg-Wilson Hamiltonian as defined and studied by Hornreich *et al.*[2] Therefore, the Lifshitz point in MnP should exhibit the characteristic critical behavior of the uniaxial one-component case, as it is suggested by the recent experimental results.

To summarize, our interpretation of the Lifshitz point in MnP is in agreement with the known experimental facts about this magnetic compound, and in particular supports the conclusion of Becerra *et al.* that its "triple point" is indeed a uniaxial one-component Lifshitz point. We intend to publish elsewhere a fuller account of the calculations outlined here.

Finally, we would like to thank Dr. Carlos C. Becerra for very useful discussions.

[1]C. C. Becerra, Y. Shapira, N. F. Oliveira, Jr., and T. S. Chang, Phys. Rev. Lett. 44, 1692 (1980).
[2]R. M. Hornreich, M. Luban, and S. Shtrikman, Phys. Rev. Lett. 35, 1678 (1975). See also R. M. Hornreich, J. Magn. Magn. Mater. 15–18, 387 (1980).
[3]D. Mukamel, J. Phys. A 10, L249 (1977); R. M. Hornreich and A. D. Bruce, *ibid.* 11, 595 (1978).
[4]A. Michelson, Phys. Rev. B 16, 577 (1977); S. Redner and H. E. Stanley, J. Phys. C 10, 4765 (1977); Phys. Rev. B 16, 4901 (1977); W. Selke, Z. Phys. B 29, 133 (1978).
[5]M. E. Fisher and W. Selke, Phys. Rev. Lett. 44, 1502 (1980). See also W. Selke and M. E. Fisher, Phys. Rev. B 20, 257 (1979).
[6]See, however, the proposals of a Lifshitz point in the field-temperature phase diagram of liquid crystals by A. Michelson, Phys. Rev. Lett. 39, 464 (1977), and by R. Blinc and F. C. S. Barreto, Phys. Status Solidi B 87, K105 (1978).
[7]C. S. O. Yokoi, M. D. Coutinho-Filho, and S. R. Salinas, Phys. Rev. B (in press).
[8]See, for example, A. Takase, H. Yashima, and T. Kasuya, J. Phys. Soc. Jpn. 47, 531 (1979).
[9]S. Hiyamizu and T. Nagamiya, Intern. J. Magn. 2, 33 (1972).
[10]See, for example, the neutron diffraction studies by G. P. Felcher, J. Appl. Phys. 37, 1056 (1966); Y. Ishikawa, T. Komatsubara, and E. Hirahara, Phys. Rev. Lett. 23, 532 (1969); H. Obara, Y. Endoh, Y. Ishikawa, and T. Komatsubara, J. Phys. Soc. Jpn. 49, 928 (1980).
[11]Y. Kitano and T. Nagamiya, Prog. Theor. Phys. 31, 1 (1964).
[12]K. Tajima, Y. Ishikawa, and H. Obara, J. Magn. Magn. Mater. 15–18, 373 (1980). See also Obara, Endoh, Ishikawa, and Komatsubara, Ref. 10.
[13]R. M. Moon, J. W. Cable, and Y. Shapira, J. Appl. Phys. (in press).
[14]See Michelson, Ref. 4
[15]Y. Shapira, C. C. Becerra, N. F. Oliveira, Jr., and T. S. Chang (unpublished).
[16]D. R. Nelson and M. E. Fisher, Phys. Rev. B 11, 1030 (1975).