Spin-localized model for the Lifshitz point in MnP

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We present a theoretical interpretation for the occurrence of a Lifshitz point (LP) in the field-temperature (H-T) phase diagram of MnP. On the basis of a simple spin-localized model, with the assumption that the exchange constants depend on H and T, we calculate the thermodynamic properties of MnP asymptotically close to the LP. In particular, we determine asymptotic expressions for the transition lines which meet tangentially at the LP. The predictions of the model concerning the behavior of the uniform transverse and longitudinal susceptibilities, and some other thermodynamic quantities, are in agreement with the reported experimental data. Finally, a renormalizaton-group analysis of the model shows that near the LP the Hamiltonian assumes essentially the form of the uniaxial ($m=1$), one component ($n=1$), Landau-Ginzburg-Wilson Hamiltonian suitable to describe a LP in magnetic systems with uniaxial symmetry. All of these results are in agreement with the suggestion that MnP has a triple point, where paramagnetic, ferromagnetic, and helicoidal (fan) phases meet, with the characteristic properties of a uniaxial one-component LP.

I. INTRODUCTION

The novel multicritical Lifshitz point (LP) (Ref. 1), which divides the phase diagram into modulated, disordered, and uniformly ordered phases, has been searched for in a variety of real physical systems, including magnetic compounds and alloys,[2,3] liquid crystals,[4] charge-transfer salts,[5] and systems exhibiting structural phase transitions.[6] The first successful attempt to find this type of multicritical behavior has been reported by Becerra *et al.*,[2] who give experimental evidence that the triple point of MnP, where paramagnetic, ferromagnetic, and helicoidal (fan) phases meet, shows the characteristic properties of a LP. In Fig. 1 we sketch the field-temperature (H-T) phase diagram of MnP in the neighborhood of the triple point. The magnetic field is applied along the $b\equiv y$ direction (intermediate axis), and the magnetic moments lay in the b-c plane ($a\equiv z$ is an extremely hard axis). In the ferromagnetic phase, in zero field, the moments point parallel to the easy axis $c\equiv x$. In the fan phase the moments rotate (but do not undergo a full rotation[7] as in a screw phase) in the b-c plane, with a propagation vector along the a axis. The more relevant experimental results for the present work are the following. (i) the crossover exponent, $\phi=0.634\pm0.03$, determined from the shape of the phase boundaries which meet tangentially at the triple point (see Fig. 1). This value of ϕ, as well as the shape of the phase boundaries, which were determined by measurements of the transverse susceptibility, χ^x, are expected on theoretical grounds[8,9] for a LP characterized by the lattice dimensionality $d=3$, a one-component order parameter, $n=1$, and a unique direction of the wave vector instability, $m=1$. Furthermore, χ^x is continuous and displays a finite cusp across the para-fan transition line $H_\lambda(T)$. As we move along $H_\lambda(T)$, χ^x diverges as $T\to T_L$, in agreement with the expected behavior at a LP. On the other hand, χ^x is discontinuous across the ferro-fan first-order transition line $H_1(T)$. (ii) The longitudinal susceptibility χ^y is constant throughout the ferromagnetic phase, and shows a discontinuous behavior across the transition lines.[2] (iii) The wave vector $\vec{q}$ which characterizes the ordering in the fan phase, determined from neutron scattering experiments,[10] goes to zero continuously as the triple point is approached. The exponent β_k, with values between 0.44 and 0.4, obtained from the temperature dependence of $\vec{q}$ along the critical line is in reasonable agreement with the theoretical results.[8] (iv) Finally, neutron spin-wave scattering experiments[11] suggest a competition between ferromagnetic and antiferromagnetic exchange interactions along the a direction. The estimated ratio of the competing exchange interactions is close to the value $\frac{1}{4}$ as in the case of the LP in the axial next-nearest-neighbor Ising (ANNNI) model.[12]

Despite the fact that the LP in MnP seems to belong to the same universality class of some simple model systems such as the ANNNI model, it nevertheless presents some remarkable particularities. For instance, since Lifshitz points are usually associated with T-p 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 H-T phase diagrams. In this respect we mention that the ANNNI model does not exhibit a Lifshitz point in the H-T phase

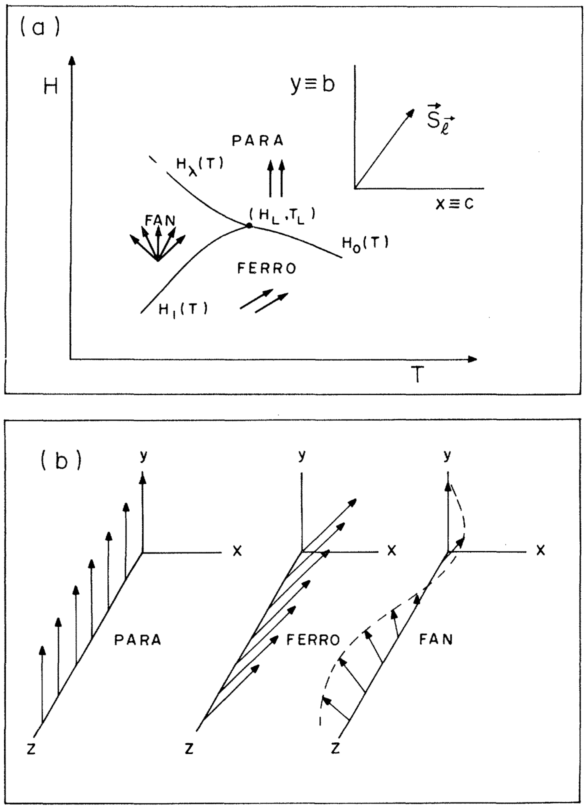

FIG. 1. (a) Schematic representation of the field-temperature phase diagram of MnP in the neighborhood of the "triple point." The phase boundaries meet tangentially at (H_L,T_L). $H_\lambda(T)$ and $H_0(T)$ are lines of critical points. $H_1(T)$ is a line of first-order transitions. The magnetic field is applied along the $b \equiv y$ direction, and the moments are on the b-c plane. The propagation vector in the fan phase is along the $a \equiv z$ direction. (b) Sketch 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 an oscillating behavior along the z direction.

diagram.[13] 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 should be suitable for this magnetic crystal.

In this work we report a possible interpretation for the experimental results of Becerra *et al.*[2] 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,[14] we remark that a localized spin model has been successfully applied to explain many magnetic properties of MnP (Ref. 15). A preliminary report based on the mean-field (MF) approximation has already been published.[16] The present paper contains a detailed account of these results, as well as a renormalization-group (RG) analysis which shows that the Hamiltonian used to describe MnP belongs to the universality class of the uniaxial $(m=1)$ one-component $(n=1)$ Landau-Ginzburg-Wilson Hamiltonian as defined and studied by Hornreich *et al.*[1] The statistic mechanical calculations in the MF approximation, together with a RG analysis, give results in qualitative agreement with the above-mentioned experimental features, and do support the suggestion concerning the nature of the triple point.

The outline of the present paper is as follows. In Sec. II we introduce the model system and obtain the relevant expressions in the MF approximation. In Sec. III we use these results to examine the conditions under which there is the occurrence of a LP in this system. In particular, asymptotic expressions for the transition lines are obtained. The predictions of the model concerning the uniform transverse and longitudinal susceptibilities are discussed in Sec. IV. These results are then compared with typical experimental data for MnP. In Sec. V we present a RG analysis of the model Hamiltonian.

II. MODEL SYSTEM AND MEAN-FIELD EQUATIONS

Hiyamizu and Nagamiya[15] have interpreted the magnetization process in MnP on the basis of a spin-localized model which takes into account the competing exchange interactions along the a axis and the spin anisotropies. Since our aim is directed towards the understanding of the phenomenon rather than to the quantitative analysis of experimental results, we assume a simple model Hamiltonian, and disregard those features of the system which are not essential to the present work. More specifically, we suppress single-ion anisotropies and write the X-Y anisotropic Hamiltonian,

$$\mathcal{H} = -\tfrac{1}{2}\sum_{\alpha}\sum_{\vec{\ell},\vec{\ell}\,'} J^{\alpha}(\vec{\ell}-\vec{\ell}\,')S^{\alpha}_{\vec{\ell}}S^{\alpha}_{\vec{\ell}\,'} - H\sum_{\vec{\ell}} S^{y}_{\vec{\ell}} , \quad (2.1)$$

where $\vec{\ell} \equiv (l,m,n)$ is a lattice vector of a simple cubic lattice with N sites in each direction, and $\vec{S}_{\ell} \equiv (S^x_{\vec{\ell}}, S^y_{\vec{\ell}})$ are Pauli spin-$\frac{1}{2}$ operators. The exchange parameters are given by

$$J^{\alpha}(\vec{\ell}-\vec{\ell}\,') = J(\vec{\ell}-\vec{\ell}\,') \pm D(\vec{\ell}-\vec{\ell}\,')\delta_{n,n'} , \quad (2.2)$$

where $\alpha = x,y$, and the plus (minus) sign holds for $\alpha = x$ (y). The interactions should be ferromagnetic in the planes $(n=n')$ and competitive along the z direction.

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 a similar X-Y Hamiltonian had already been subjected to a theoretical investigation by Kitano and Nagamiya[17] two decades ago, but of course they had not focused their attention on the problem of the LP.

The MF expression for the Gibbs free energy $G(T,H,N)$ of this system may be derived via the inequality

$$G \le \Phi = G_0 + \langle \mathcal{H} - \mathcal{H}_0 \rangle_0 , \quad (2.3)$$

where

$$G_0=-kT\ln[\mathrm{Tr}\exp(-\beta\mathscr{H}_0)]\ ,\tag{2.4}$$

$\beta=1/kT$, $\mathscr{H}_0$ is a trial Hamiltonian, the trace is over spin configurations, and the average $\langle\mathscr{H}-\mathscr{H}_0\rangle_0$ is taken with respect to $\mathscr{H}_0$. To obtain the MF approximation, we consider the free trial Hamiltonian

$$\mathscr{H}_0=-\sum_{\vec{l}}\vec{\eta}_n\cdot\vec{S}_{\vec{l}}\ ,\tag{2.5}$$

where $\vec{\eta}_n$ is a variational parameter associated with the nth layer. It then follows the MF Gibbs free energy

$$G_{\mathrm{MF}}(T,H,N)=\min_{\{\vec{M}_n\}}\Phi(T,H,N;\{\vec{M}_n\})\ ,\tag{2.6}$$

where

$$N^{-3}\Phi(T,H,N;\{\vec{M}_n\})=-kT\ln 2+\frac{kT}{N}\sum_n\int_0^{M_n}\tanh^{-1}\mu\,d\mu-\frac{1}{2N}\sum_{n,n'}J(n-n')\vec{M}_n\cdot\vec{M}_{n'}$$
$$-\frac{D}{N}\sum_n[(M_n^x)^2-(M_n^y)^2]-\frac{H}{N}\sum_n M_n^y\ .\tag{2.7}$$

In this last equation the effective isotropic exchange constants are given by

$$J(n-n')=N^{-2}\sum_{l,m}\sum_{l',m'}J(l-l',m-m',n-n')\ ,\tag{2.8}$$

while the effective anisotropic exchange constant is

$$2D=N^{-2}\sum_{l,m}\sum_{l',m'}D(l-l',m-m',0)\ .\tag{2.9}$$

The average layer magnetization $\vec{M}_n\equiv(M_n^x,M_n^y)$ is given by the minimization conditions

$$M_n^\alpha=\frac{\eta_n^\alpha}{\eta_n}M_n\ ,\tag{2.10}$$

where

$$M_n=[(M_n^x)^2+(M_n^y)^2]^{1/2}\ ,$$
$$\eta_n=[(\eta_n^x)^2+(\eta_n^y)^2]^{1/2}\ ,\tag{2.11}$$
$$M_n=\tanh\beta\eta_n\ ,$$

and

$$\eta_n^\alpha=\sum_{n'}J(n-n')M_{n'}^\alpha\pm 2DM_n^\alpha+H_n^\alpha\ ,\tag{2.12}$$

where $\alpha=x,y$, the plus (minus) sign holds for $\alpha=x$ (y), and $H_n^x=0$, $H_n^y=H$.

III. TRANSITION LINES

In this section we will examine, within the MF approximation, the shape of the transition lines which are expected to meet tangentially at the LP.

A. Transverse paramagnetic susceptibility and second-order transition lines

The paramagnetic phase is specified by a uniform magnetization, $\vec{M}_n=(0,M)$, along the b (y) axis. From Eqs. (2.8)–(2.11), M satisfies the equation

$$M=\tanh\beta\{[\hat{J}(0)-2D]M+H\}\ ,\tag{3.1}$$

where

$$\hat{J}(q)=\sum_n J(n)e^{iqn}\ .\tag{3.2}$$

The transition from the paramagnetic phase to the ordered phases, either ferromagnetic or fan, is characterized by the onset of a nonzero x component of the magnetization. It is then of interest to calculate the wave-vector-dependent transverse susceptibility $\chi^x(q)$ by considering a perturbation field $H_n^x=\delta h_n^x$ in Eq. (2.12). Using Eqs. (2.10)–(2.12) we obtain

$$\tfrac{1}{2}kT\frac{\delta M_n^x}{M_n}\ln\left[\frac{1+M_n}{1-M_n}\right]=\sum_{n'}J(n-n')\delta M_{n'}^x+2D\delta M_n^x+\delta h_n^x\ ,\tag{3.3}$$

and

$$\tfrac{1}{2}kT\frac{M_n^y}{M_n}\ln\left[\frac{1+M_n}{1-M_n}\right]=\sum_{n'}J(n-n')M_{n'}^y-2DM_n^y+H\ .\tag{3.4}$$

As the susceptibility is calculated in the limit $\delta h_n^x\to 0$, which implies $M_n\to M_n^y=M$, this system of equations may be written in the form

$$\{[\hat{J}(0)-2D]M+H\}\frac{\delta M_n^x}{M}=\sum_{n'}J(n-n')\delta M_{n'}^x+2D\delta M_n^x+\delta h_n^x\ .\tag{3.5}$$

By defining the Fourier transforms

$$\delta M_n^x=\sum_q e^{-iqn}\delta m_q^x\ ,\tag{3.6}$$

$$\delta h_n^x=\sum_q e^{-iqn}\delta h_q^x\ ,\tag{3.7}$$

the wave-vector-dependent paramagnetic transverse susceptibility $\chi_p^x(q)=\delta m_q^x/\delta h_q^x$, is found to be given by

$$\chi_p^x(q)=[\hat{J}(0)-\hat{J}(q)-4D+M^{-1}H]^{-1}\ .\tag{3.8}$$

The transition to the ordered phases is characterized by the divergence of $\chi_p^x(q)$, namely, by the condition

$$\hat{J}(0)-4D+M^{-1}H=\max_q\hat{J}(q)=\hat{J}(q_c)\ .\tag{3.9}$$

From this last equation, and since $\hat{J}(q)$ 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 H-T phase diagram near the border of the paramagnetic phase. However, it is an experimental fact that both phases are present. Therefore, in the framework of the MF approximation, there is no alternative but to impose the *ad hoc* dependence of $\hat{J}(q)$ on 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.[11] In any event, Eq. (2.7) should be regarded as an effective free energy in which these effects have already been taken into account. $\hat{J}(q)$ may thus be expanded about $q=0$ in the form

$$\hat{J}(q)=\hat{J}(0)-\alpha q^2-\tfrac{1}{2}\beta q^4-\cdots , \tag{3.10}$$

where, due to the occurrence of the Lifshitz point (T_L,H_L) for $\alpha=0$, we write in leading order

$$\alpha=\alpha_T(T-T_L)+\alpha_H(H-H_L) , \tag{3.11}$$

$$\hat{J}(0)=J_0+J_{0T}(T-T_L)+J_{0H}(H-H_L) . \tag{3.12}$$

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 LP.

If $\alpha>0$, the maximum of $\hat{J}(q)$ occurs for $q=0$, and the system undergoes a second-order para-ferromagnetic transition. The transition line is determined by Eqs. (3.1) and (3.9), for $q_c=0$, and is given by

$$\frac{H}{4D}=\tanh\frac{H[\hat{J}(0)+2D]}{4kTD} . \tag{3.13}$$

By using Eq. (3.12) it is possible to show that the para-ferro transition line, $H_0(T)$, has the asymptotic form

$$H_0(T)\approx H_L-A(\Delta T)-B(\Delta T^2) , \tag{3.14}$$

where $\Delta T=T-T_L$, and A and B are given in Appendix A. On the other hand, if $\alpha<0$, the maximum of $\hat{J}(q)$ occurs for

$$q_c=(-\alpha/\beta)^{1/2} , \tag{3.15}$$

that is, the state of equilibrium in the ordered phase is spatially nonuniform and there is a second-order para-fan transition. From Eqs. (3.1) and (3.9) we obtain, for $q_c=(-\alpha/\beta)^{1/2}$, the transition line

$$\frac{H}{\hat{J}(q_c)-\hat{J}(0)+4D}=\tanh\frac{[\hat{J}(q_c)+2D]H}{[\hat{J}(q_c)-\hat{J}(0)+4D]kT} . \tag{3.16}$$

Near the LP the asymptotic form of $H_\lambda(T)$, which may be obtained using Eq. (3.11), with $\hat{J}(q_c)=\hat{J}(0)+\alpha^2/2\beta$, is given by

$$H_\lambda(T)\approx H_0(T)+C(\Delta T)^2 , \tag{3.17}$$

where C is defined in Appendix A.

From Eq. (3.17), it is apparent that both critical lines meet tangentially at the Lifshitz point, and that the MF scaling crossover exponent is $\phi=\frac{1}{2}$. These lines are drawn schematically in Fig. 1. We may also conclude, from Eqs. (3.15) and (3.17), that on the para-fan transition line the wave vector q_c behaves asymptotically as

$$q_c^2\propto(T-T_L)\propto(H-H_L) . \tag{3.18}$$

This result, which implies the MF value $\frac{1}{2}$ for the exponent β_k, is in fairly close agreement with the neutron-diffraction data.[10] Also, RG calculations[1,8] predict $\beta_k=\frac{1}{2}$ to order ϵ^2.

B. Landau expansion and first-order ferro-fan transition line

In order to determine the ferro-fan transition line we have to compare the Gibbs free energy in both phases. This will be done by considering a Landau expansion for the free energy. Defining $\vec{M}_n=(\delta M_n^x, M+\delta M_n^y)$, with M given by (3.1), the MF Gibbs functional (2.7) may be expanded in the form

$$\begin{aligned}N^{-3}[\Phi(T,H,N;\{\delta M_n^x,\delta M_n^y\})-G_0]=&-\frac{1}{2N}\sum_{n,n'}J(n-n')(\delta M_n^x\delta M_{n'}^x+\delta M_n^y\delta M_{n'}^y)\\&+\frac{1}{N}\sum_n[(A_{20}-D)\delta M_n^{x^2}+(A_{02}+D)\delta M_n^{y^2}+A_{21}\delta M_n^{x^2}\delta M_n^y+A_{03}\delta M_n^{y^3}\\&+A_{40}\delta M_n^{x^4}+A_{04}\delta M_n^{y^4}+A_{22}\delta M_n^{x^2}\delta M_n^{y^2}+\cdots] ,\end{aligned} \tag{3.19}$$

where G_0, the free energy in the paramagnetic phase, and the coefficients A_{ij} are given in Appendix B.

In the ferromagnetic phase $\vec{M}_n$ is uniform, that is, $\delta M_n^x\equiv\delta M^x$, $\delta M_n^y\equiv\delta M^y$. By minimizing Φ with respect to δM^y, we get in leading order

$$\delta M^y\approx-\frac{A_{21}}{2[A_{02}+D-\frac{1}{2}\hat{J}(0)]}\delta M^{x^2} . \tag{3.20}$$

We thus obtain the Gibbs functional up to fourth order in δM^x,

$$N^{-3}(\Phi^{(f)}-G_0)\approx A(0)\delta M^{x^2}+\left[A_{40}-\frac{A_{21}^2}{4B(0)}\right]\delta M^{x^4} , \tag{3.21}$$

where

$$A(q)=A_{20}-D-\tfrac{1}{2}\hat{J}(q)=[2\chi_p^x(q)]^{-1} \tag{3.22}$$

and

$$B(q)=A_{02}+D-\tfrac{1}{2}\hat{J}(q) . \tag{3.23}$$

This form of the free energy will be useful to determine the transverse uniform susceptibility in the ferro phase, χ_F^x in the next section. The minimization of Φ gives the value of δM^x (in the ferromagnetic phase) near the paraferro transition line,

$$\delta M^x = \pm \left[-\frac{A(0)}{2\left[A_{40} - \frac{A_{21}^2}{4B(0)}\right]} \right]^{1/2}$$

$$\propto \pm |H - H_0(T)|^{1/2} . \quad (3.24)$$

This last expression confirms the usual mean-field critical exponent $\beta = \frac{1}{2}$. The leading asymptotic expression for the free energy in the ferromagnetic phase is given by

$$N^{-3}[G_{\rm MF}^{(f)}(T,H,N) - G_0] = -\frac{A^2(0)}{4A_{40} - \frac{A_{21}^2}{B(0)}} . \quad (3.25)$$

In the fan phase, $\vec{M}_n$ is spatially nonuniform (modulated), and the derivation of a Landau expansion in terms of the order parameter is more complex. It is convenient to introduce the Fourier transform

$$\delta M_n^y = \sum_q e^{-iqn} \delta m_q^y . \quad (3.26)$$

By minimizing the free energy (3.19) with respect to δM_n^y we obtain

$$\delta m_q^y = -\frac{A_{21}}{2B(q)} \sum_{q'q''} \delta m_{q'}^x \delta m_{q''}^x \Delta(q'+q''-q) + \cdots , \quad (3.27)$$

where δm_q^x was defined by Eq. (3.6) and the Δ function, given by

$$\Delta(q) = \sum_{n=-\infty}^{\infty} \delta_{q,2\pi n} , \quad (3.28)$$

expresses the wave-vector conservation modulo 2π. We can thus write the expansion of Φ in the form

$$N^{-3}[\Phi(T,H,N;\{\delta m_q^x\}) - G_0] = \sum_{q_1 q_2} A(q_1) \delta m_{q_1}^x \delta m_{q_2}^x \Delta(q_1+q_2) - \sum_q \frac{A_{21}^2}{4B(q)} \left[\sum_{q_1 q_2} \delta m_{q_1}^x \delta m_{q_2}^x \Delta(q_1+q_2+q) \right] \left[\sum_{q_1 q_2} \delta m_{q_1}^x \delta m_{q_2}^x \Delta(q_1+q_2-q) \right] + A_{40} \sum_{q_1 q_2 q_3 q_4} \delta m_{q_1}^x \delta m_{q_2}^x \delta m_{q_3}^x \delta m_{q_4}^x \Delta(q_1+q_2+q_3+q_4) + \cdots . \quad (3.29)$$

As in the sinusoidal (Ising) case,[13] let us define

$$\delta m_0^x = M_0^x , \quad 2\delta m_{nq_c}^x = M_n^x e^{i\phi} \quad (n \ge 1) , \quad (3.30)$$

and search for a solution in the form

$$\delta M_n^x = M_0^x + M_1^x \cos(q_c + \phi) + M_2^x \cos 2(q_c + \phi) + M_3^x \cos 3(q_c + \phi) + \cdots , \quad (3.31)$$

where $M_0^x, M_1^x, M_2^x, \ldots$, are real numbers and ϕ is an arbitrary phase. Since umklapp terms near the LP do not contribute in lower orders (see the discussion in Appendix A of Ref. 13), we minimize the free energy with respect to M_n^x and obtain

$$M_n^x = 0 \ (\text{for } n \text{ even}) , \qquad M_n^x \propto (M_1^x)^n \ (\text{for } n \text{ odd}) . \quad (3.32)$$

On the other hand, using a Fourier expansion for δM_n^y,

$$\delta M_n^y = M_0^y + M_1^y \cos(q_c + \phi) + M_2^y \cos[2(q_c + \phi)] + M_3^y \cos[3(q_c + \phi)] + \cdots , \quad (3.33)$$

we obtain

$$M_n^y = 0 \ (\text{for } n \text{ odd}) , \qquad M_0^y \propto (M_1^x)^2 , \quad M_n^y \propto (M_1^x)^n \ (\text{for } n \text{ even}, n \ge 2) . \quad (3.34)$$

These results imply that the leading asymptotic expression of Φ is determined by the main harmonic component of δM_n^x, that is,

$$N^{-3}[\Phi^{(m)}(T,H,N;M_1^x) - G_0] \approx \tfrac{1}{2} A(q_c)(M_1^x)^2 + \frac{1}{16}\left[6A_{40} - \frac{A_{21}^2}{B(0)} - \frac{A_{21}^2}{2B(2q_c)} \right] (M_1^x)^4 . \quad (3.35)$$

By minimizing $\Phi^{(m)}$ with respect to M_1^x, we obtain

$$M_1^x = \pm 2 \left[\frac{-A(q_c)}{6A_{40} - A_{21}^2 \left[\frac{1}{B(0)} + \frac{1}{2B(2q_c)} \right]} \right]^{1/2}$$

$$\propto \pm |H - H_\lambda(T)|^{1/2} . \quad (3.36)$$

Thus the leading asymptotic expression for the free energy in the fan (modulated) phase is given by

$$N^{-3}[G_{\rm MF}^{(m)}(T,H,N) - G_0] = -\frac{A^2(q_c)}{6A_{40} - A_{21}^2 \left[\frac{1}{B(0)} + \frac{1}{2B(2q_c)} \right]} . \quad (3.37)$$

Now, by comparing the free energies (3.25) and (3.37), and using Eqs. (3.10)–(3.12), we obtain the following asymptotic expression for the ferro-fan transition line close to the LP:

$$H_1(T) \approx H_0(T) - C(2+\sqrt{6})\Delta T^2 \ . \tag{3.38}$$

Therefore, in agreement with the experimental data, the three transition lines meet tangentially at the LP. It is interesting to observe that the ratio $[H_0(T) - H_1(T)]/[H_\lambda(T) - H_0(T)] = \sqrt{6}+2$, which was first noticed by Michelson[9] in a similar but different context, still holds in the present model. 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 MF approximation.

IV. UNIFORM TRANSVERSE AND LONGITUDINAL SUSCEPTIBILITIES

The phase diagram of MnP was determined from magnetostriction and differential susceptibility measurements, as discussed in detail by Shapira *et al.*[2] Therefore, it is of interest to examine the predictions of the model concerning the behavior of the uniform transverse and longitudinal susceptibilities, particularly in the vicinity of the LP. The derivations will be based on the MF results presented in the previous two sections.

A. Uniform transverse susceptibility

In the paramagnetic phase the uniform transverse susceptibility, $\chi_p^x(q=0) \equiv \chi_p^x$, is obtained from Eq. (3.8) for $q=0$,

$$\chi_p^x = \frac{M}{H-4DM} \ , \tag{4.1}$$

where M is determined by Eq. (3.1). At constant T, and near the para-ferro transition line, we can write the asymptotic expression

$$\chi_p^x = \frac{C_0}{H-H_0(T)} \ , \tag{4.2}$$

where

$$C_0 = \left[\frac{M^2}{M - H\dfrac{\partial M}{\partial H}} \right]_{T,H_0(T)} \ . \tag{4.3}$$

Therefore, χ_p^x obeys the usual Curie-Weiss law across the $H_0(T)$ transition line with an associated MF exponent $\gamma=1$.

On the other hand, by considering a perturbation field δh^x, which implies an additional term $(-\delta h^x \delta M^x)$ in Eq. (3.21), we can find, after minimization, the uniform transverse susceptibility in the ferromagnet phase, $\chi_F^x = \delta M^x/\delta h^x$,

$$\chi_f^x = \frac{-M}{2(H-4DM)} \ . \tag{4.4}$$

Near the para-ferro transition line we have the asymptotic form

$$\chi_f^x \approx \frac{C_0}{2[H_0(T)-H]} \ . \tag{4.5}$$

In order to calculate χ^x in the fan phase, we have to consider the uniform component M_0^x of δM_n^x, which is coupled to the uniform perturbation field δh^x. By adding

$$\tfrac{1}{2}(\chi_p^x)^{-1} M_0^{x\,2} + B_{10} M_0^{x\,2} M_1^{x\,2} - \delta h^x M_0^x$$

to the free-energy functional (3.35), minimizing it with respect to M_0^x, and using (3.36), we can derive the uniform transverse susceptibility in the fan phase,

$$\chi_m^x = \frac{1}{[\chi_p^x(0)]^{-1} - \mathscr{M}[\chi_p^x(q_c)]^{-1}} \ , \tag{4.6}$$

where $\chi_p^x(q)$ is given by Eq. (3.8) and $\mathscr{M}$ is defined in Appendix C. In the vicinity of the Lifshitz point, we can write

$$\chi_m^x \approx \frac{C_0}{[H-H_0(T)] - C'[H-H_\lambda(T)]} \ , \tag{4.7}$$

where C' is given in Appendix C. From the results derived above we conclude that χ^x is continuous and shows a finite cusp across the para-fan transition line $H_\lambda(T)$. As we move along the para-fan line, χ^x diverges as $|\Delta T|^{-2}$ for $T \to T_L$. On the other hand, χ^x is discontinuous across the ferro-fan first-order transition line $H_1(T)$. All these features are in qualitative agreement with the experimental measurements.

In comparing the theoretical predictions for χ^x with the experimental data,[2] we have to consider the demagnetization effects, particularly in the transition on the para-ferro boundary. The transition on this boundary is associated with a divergence in the intrinsic susceptibility [Eqs. (4.2) and (4.5)]. However, because of the demagnetization effects, the measured average susceptibility on this boundary has the upper limit $\bar{\chi}_{me} = 1/N$, where N is the demagnetization factor of the ellipsoidal sample. In the ferro phase, the ferromagnetic domains keep $\bar{\chi}_{me}$ with the value $1/N$. The main point to observe is that in our theoretical discussion the field H is the internal field $H = H_0 - NM$, where H_0 is the external field applied along one of the principal axes of the ellipsoid (see Shapira *et al.*[2] for details). In Figs. 2 and 3 we sketch the predicted MF behavior for χ^x as a function of the internal field H, and compare it with sketches of typical data,[2] for both $T < T_L$ and $T > T_L$, as a function of the applied field H_0. In analogy to the case of simple ferromagnets, the discrepancies are explained by the role of the demagnetization corrections (see Shapira *et al.*[2] for a detailed discussion of this point).

B. Uniform longitudinal susceptibility

In the paramagnetic phase $M_n^x = 0$ and $M_n^y = M$. Thus the parallel susceptibility $\chi^y - \partial M^y/\partial H^y$ may be evaluated from (3.1) and (3.12),

$$\chi_p^y = \frac{1+J_{0H}}{kT(1-M^2)^{-1} + 2D - \hat{J}(0)} \ . \tag{4.8}$$

On the other hand, in the ferromagnetic phase we have,

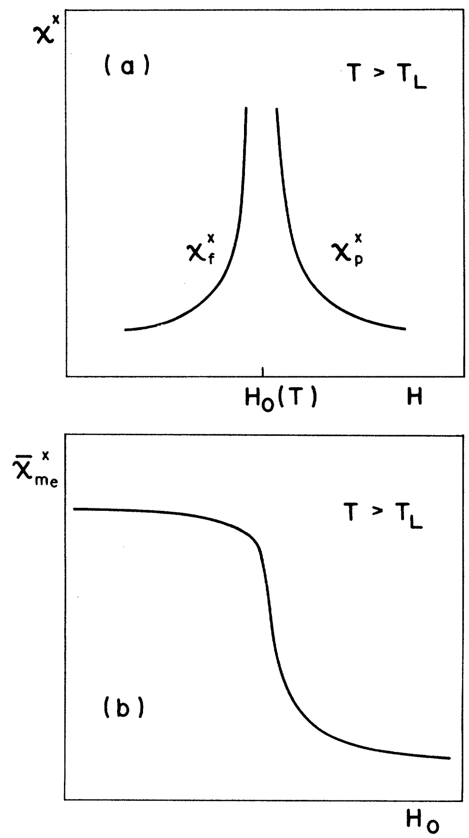

FIG. 2. (a) Sketch of the predicted MF behavior for the uniform transverse susceptibility χ^x near the para-ferro second-order transition $H_0(T)$ as a function of the internal field H. (b) Sketch of typical experimental data (from Ref. 2) for the average uniform transverse susceptibility, $\bar{\chi}^x_{me}$, near the para-ferro transition as a function of the applied field H_0. Owing to demagnetization effects, $\bar{\chi}^x_{me}$ is bounded by an upper limit N^{-1}, where N is the demagnetization factor of the ellipsoidal sample. In the ferro phase the ferromagnetic domains keep $\bar{\chi}^x_{me}$ with the fixed value N^{-1}.

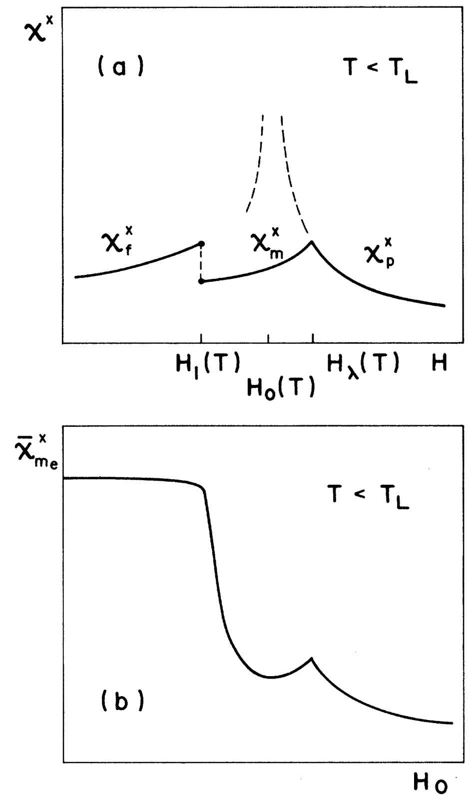

FIG. 3. (a) Sketch of the predicted MF behavior of the uniform transverse susceptibility χ^x near the second-order para-fan, $H_\lambda(T)$, and first-order fan-ferro, $H_1(T)$, transitions as a function of the internal field H. The dashed lines illustrate the divergence of χ^x near the para-ferro transition, $H=H_0$, when $T>T_L$ [see Fig. 2(a)]. (b) Sketch of typical data (from Ref. 2) for the average uniform transverse susceptibility $\bar{\chi}^x_{me}$ near the para-fan and fan-ferro transitions, as a function of the applied field H_0. As in Fig. 2(b), in the ferro phase the ferromagnetic domains keep $\bar{\chi}^x_{me}$ with the constant value N^{-1}.

from Eqs. (2.10)—(2.12), the set of equations

$$\tfrac{1}{2}kT\ln\left[\frac{1+M}{1-M}\right]\frac{M^x}{M}=[\hat{J}(0)+2D]M^x\ , \qquad (4.9)$$

and

$$\tfrac{1}{2}kT\ln\left[\frac{1+M}{1-M}\right]\frac{M^y}{M}=[\hat{J}(0)-2D]M^y+H\ , \qquad (4.10)$$

which imply the relation $H=4DM^y$, or

$$\chi^y_f=\frac{1}{4D}\ . \qquad (4.11)$$

In order to determine χ^y in the fan phase, but near the $H_\lambda(T)$ line, we must resort to the Landau theory presented in the preceding section. It was shown that the uniform component M^y_0 of δM^y_n behaves asymptotically as $M^y_0\propto(M^x_1)^2\ \propto(H_\lambda-H)\ +O(H_\lambda-H)^2$. Since $M^y_n=M+\delta M^y_n$, we obtain, at constant T,

$$(\chi^y_m-\chi^y_p)_\lambda=\left[\frac{A_{21}}{4B(0)}\frac{1}{6A_{40}-A^2_{21}\left[\dfrac{1}{B(0)}+\dfrac{1}{2B(2q_c)}\right]}\frac{\partial A(q_c)}{\partial H}\right]_{T,H_0(\lambda)}\ , \qquad (4.12)$$

which means that χ^y has a temperature-dependent discontinuity across the para-fan transition line. A similar analysis, near the ferro-fan transition line, shows that χ^y is also discontinuous across the $H_1(T)$ first-order line.

Therefore, the longitudinal susceptibility χ^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.[2] Sketches of the predicted MF behavior for χ^y are shown in Figs. 4 and 5. For comparison, we also sketch typical data,[2] for both $T<T_L$ and $T>T_L$, as a function of the applied field H_0. In the latter curves, the second-order ferro-para transition is characterized by a "shoulder," while the first-order ferro-fan transition is signaled by a peak in the susceptibility curve.[2] Similar discrepancies between the experimental and the theoretical curves are known to be found in the case of small anisotropy antiferromagnets (see, for example, the discussion about the field behavior of antiferromagnets in de Jongh and Miedema[18]).

C. Further suggestions

We may 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

$$M^y_{\text{ferro}}-M^y_{\text{fan}}\propto(T-T_L)^2\propto(H-H_L)^2\ . \tag{4.13}$$

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 minimization of the Gibbs functional we obtain 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.

V. RENORMALIZATION-GROUP ANALYSIS

In this section we use RG techniques to analyze the simplified Hamiltonian (2.1) which preserves the symmetries of the original Hiyamizu and Nagamiya model for MnP. The main goal is to show that near the LP the critical properties of this X-Y Hamiltonian belong to the same universality class of the uniaxial one-component Landau-Ginzburg-Wilson Hamiltonian.

The effective Hamiltonian associated with (2.1) is

$$\overline{\mathscr{H}}\{\vec{S}_{\vec{l}}\}=\beta\mathscr{H}\{\vec{S}_{\vec{l}}\}+\sum_{\vec{l}} w\{\,|\vec{S}_{\vec{l}}|\,\}\ , \tag{5.1}$$

where the spin weighting factor is defined by

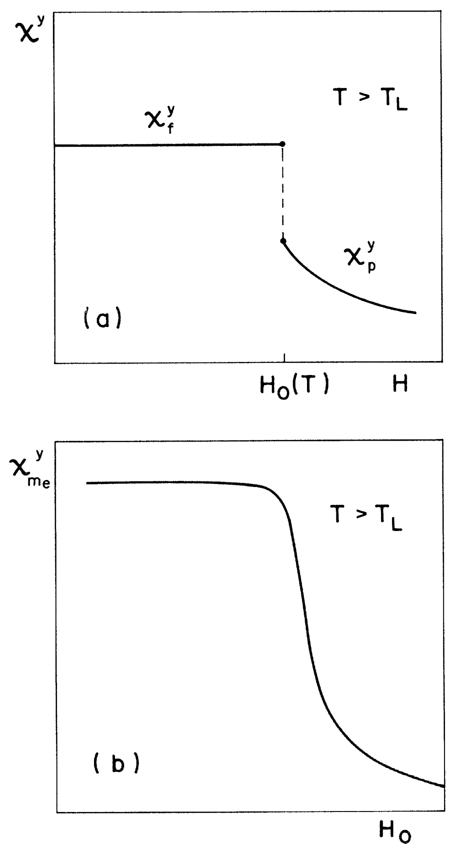

FIG. 4. (a) Sketch of the predicted MF behavior of the uniform longitudinal susceptibility χ^y near the second-order para-ferro transition $H_0(T)$ as a function of the internal field H. It should be noticed that χ^y is constant $(=1/4D)$ throughout the ferromagnetic phase. (b) Sketch of typical data (from Ref. 2) for the average uniform longitudinal susceptibility $\bar{\chi}^y_{me}$ near the para-ferro transition as a function of the external field H_0.

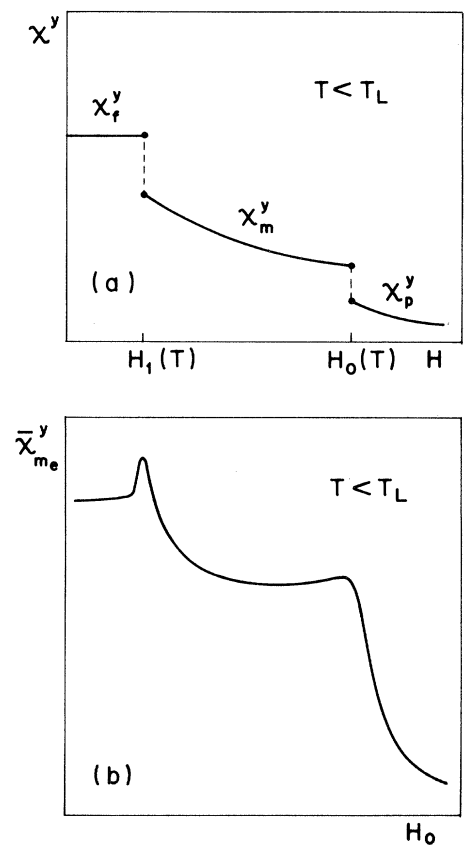

FIG. 5 (a) Sketch of the predicted MF behavior of the uniform longitudinal susceptibility χ^y near the second-order para-fan, $H_\lambda(T)$, and first-order fan-ferro, $H_1(T)$, transitions as a function of the internal field H. (b) Sketch of typical data (from Ref. 2) for the average uniform longitudinal susceptibility χ^y near the para-fan and fan-ferro transitions as a function of the external field H_0. The second-order para-ferro transition is characterized by a "shoulder," while the first-order fan-ferro transition is identified by a sharp peak in the $\bar{\chi}^y_{me}$ curve.

$$w\{|\vec{S}|\}=\tfrac{1}{2}|\vec{S}|^2+\tfrac{1}{4}b|\vec{S}|^4\ , \quad b>0\ . \tag{5.2}$$

The partition function of the system is then given by

$$Z=\int \prod_{\vec{\ell}\alpha} dS^{\alpha}_{\vec{\ell}}\, e^{-\overline{\mathscr{H}}\{S_{\vec{\ell}}\}}\ , \tag{5.3}$$

where the spin fields are two-component classical vectors. Introducing the Fourier transform

$$S^{\alpha}_{\vec{q}}=\sum_{\ell} S^{\alpha}_{\vec{\ell}}\, e^{i\vec{q}\cdot\vec{\ell}}\ , \tag{5.4}$$

the effective Hamiltonian may be written in the form

$$\overline{\mathscr{H}}\{\vec{S}_{\vec{q}}\}=\tfrac{1}{2}\int_{\vec{q}}\sum_{\alpha=1}^{2}\bar{u}^{\alpha}_{2}(\vec{q})S^{\alpha}_{\vec{q}}S^{\alpha}_{-\vec{q}}+\frac{b}{4}\int_{\vec{q}}\int_{\vec{q}\,'}\int_{\vec{q}\,''}S^{\alpha}_{\vec{q}}S^{\alpha}_{\vec{q}\,'}S^{\beta}_{\vec{q}\,''}S^{\beta}_{-\vec{q}-\vec{q}\,'-\vec{q}\,''}-\beta H S^{y}_{0}\ , \tag{5.5}$$

where $\int_{\vec{q}}\equiv(2\pi)^{-d}\int d^{d}_{q}$, d is the dimensionality of the system, and

$$\bar{u}^{\alpha}_{2}(\vec{q})=1-\beta\hat{J}^{\alpha}(\vec{q})\ . \tag{5.6}$$

It is convenient to eliminate the linear term proportional to S^{y}_{0} in Eq. (5.5). This can be done by the shift

$$S^{y}_{\vec{q}}\rightarrow S^{y}_{\vec{q}}+M\delta(\vec{q})\ , \tag{5.7}$$

where M satisfies the relation

$$-\beta H+\bar{u}^{y}_{2}(0)M+bM^{3}=0\ . \tag{5.8}$$

Apart from a spin-independent term, $\overline{\mathscr{H}}\{S_{\vec{q}}\}$ is given by

$$\mathscr{H}\{\vec{S}_{\vec{q}}\}=\tfrac{1}{2}\int_{\vec{q}}\sum_{\alpha=1}^{2}u^{\alpha}_{2}(\vec{q})S^{\alpha}_{\vec{q}}S^{\alpha}_{-\vec{q}}+w\int_{\vec{q}}\int_{\vec{q}\,'}S^{y}_{\vec{q}}\left[\sum_{\alpha=1}^{2}S^{\alpha}_{\vec{q}\,'}S^{\alpha}_{-\vec{q}-\vec{q}\,'}\right]$$
$$+u\int_{\vec{q}}\int_{\vec{q}\,'}\int_{\vec{q}\,''}\sum_{\alpha,\beta=1}^{2}S^{\alpha}_{\vec{q}}S^{\alpha}_{\vec{q}\,'}S^{\beta}_{\vec{q}\,''}S^{\beta}_{-\vec{q}-\vec{q}\,'-\vec{q}\,''}\ , \tag{5.9}$$

where

$$u^{x}_{2}(\vec{q})=\bar{u}^{x}_{2}(\vec{q})+bM^{2}\ , \tag{5.10}$$

$$u^{y}_{2}(\vec{q})=\bar{u}^{y}_{2}(\vec{q})+3bM^{2}\ , \tag{5.11}$$

$$w=bM\ , \quad u=b/4\ . \tag{5.12}$$

Since $u^{y}_{2}(\vec{q})>u^{x}_{2}(\vec{q})$, we identify $S^{x}_{\vec{q}}$ as the critical field, while $S^{y}_{\vec{q}}$ is noncritical. We can integrate over the variables $S^{y}_{\vec{q}}$, using a diagrammatic perturbative expansion, to obtain a reduced Hamiltonian $\overline{\mathscr{H}}\{S^{x}_{\vec{q}}\}$. The vertices and the relevant diagrams are shown in Fig. 6. The spin-dependent part of the effective reduced Hamiltonian may be written in the form

$$\overline{\mathscr{H}}\{S^{x}_{\vec{q}}\}=\tfrac{1}{2}\int_{\vec{q}}u_{2}(\vec{q})S^{x}_{\vec{q}}S^{x}_{-\vec{q}}+\frac{1}{4!}\int_{\vec{q}}\int_{\vec{q}\,'}\int_{\vec{q}\,''}u_{4}(\vec{q},\vec{q}\,',\vec{q}\,'')S^{x}_{\vec{q}}S^{x}_{\vec{q}\,'}S^{x}_{\vec{q}\,''}S^{x}_{-\vec{q}-\vec{q}\,'-\vec{q}\,''}+\cdots\ , \tag{5.13}$$

where the ellipses represents sixth-order terms, and where

$$u_{2}(\vec{q})=u^{x}_{2}(\vec{q})+4u\int_{\vec{p}}[u^{y}_{2}(\vec{p})]^{-1}-96u^{2}M^{2}[u^{y}_{2}(0)]^{-1}\int_{\vec{p}}[u^{y}_{2}(p)]^{-1}$$
$$-48u^{2}\int_{\vec{p}}[u^{y}_{2}(\vec{p})]^{-1}\int_{\vec{p}}[u^{y}_{2}(\vec{p})]^{-1}[u^{y}_{2}(\vec{p}+\vec{q})]^{-1}+O(u^{3})\ , \tag{5.14}$$

$$u_{4}(\vec{q},\vec{q}\,',\vec{q}\,'')=24u-192u^{2}M^{2}[u^{y}_{2}(\vec{q}+\vec{q}\,')]^{-1}-96u^{2}\int_{\vec{p}}[u^{y}_{2}(\vec{p})]^{-1}[u^{y}_{2}(\vec{p}+\vec{q}+\vec{q}\,')]^{-1}+O(u^{3})\ . \tag{5.15}$$

A Hamiltonian of this same form has been used to study LP points of the uniaxial type.[1] In particular, if we consider the low momentum expansion of $u_2(\vec{q})$, the coefficient of q^{2}_{α}, where α indicates the direction of modulation, should vanish in order to give rise to a LP. In the present case this may be achieved by a suitable control of the parameters T and H (see Sec. III). The equivalence which we have shown clarifies the agreement between the experimental crossover exponent,[2] $\phi=0.634\pm0.03$, and the theoretical value,[8] $\phi=0.625+O(\epsilon^2)$.

VI. CONCLUSIONS

We have presented a theoretical interpretation, based on a simple spin localized model, for the occurrence of a LP in the phase diagram of MnP. In particular, we have determined asymptotic expressions for the transition lines which meet tangentially at the LP. The predictions of the model concerning the behavior of the uniform transverse and longitudinal susceptibilities and other thermodynamic quantities are in qualitative agreement with the reported experimental data.[2] Finally, a renormalization-group analysis of the model shows that near the LP the Hamiltonian assumes the form of the one-component ($n=1$) uniaxial ($m=1$) Landau-Ginzburg-Wilson Hamiltonian appropriated to describe a LP in magnetic systems with uniaxial symmetry.[1] In conclusion, our interpretation of a LP in MnP is in agreement with the known experimental facts about this compound, and in particular supports the suggestion[2] that its "triple point" is indeed a uniaxial one-component LP.

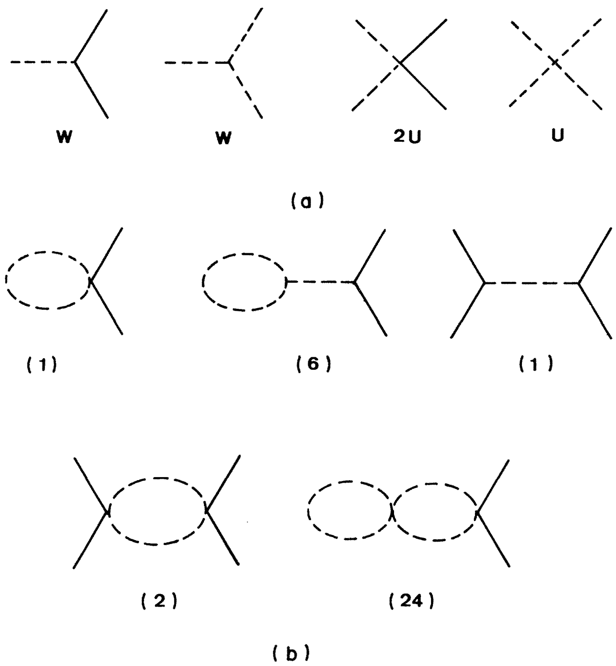

FIG. 6. (a) Perturbative vertices in the diagrammatic expansion. The dashed lines represent the variables $S^{y}_{\vec{q}}$, and the solid lines the variables $S^{x}_{\vec{q}}$. (b) Contributions, up to order u^2, to the diagrammatic perturbative expansion. The numbers indicate the multiplicity of each diagram.

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APPENDIX A

The values of the coefficients in Eqs. (3.14), (3.17), and (3.38) are the following:

$$A=\frac{a}{b}\ , \tag{A1}$$

$$B=\frac{a}{b^2}\left[\frac{ac}{b}-d\right], \tag{A2}$$

$$C=-\frac{M_L}{2\beta}\left[\frac{kT_L}{1-M_L^2}-J_0+2D\right]\times\frac{a}{b^2}\left[\frac{b}{a}\alpha_T^2+2\alpha_T\alpha_H-\frac{\alpha}{b}\alpha_H^2\right], \tag{A3}$$

$$a=H_L\left[J_{OH}-\frac{J_0+2D}{T_L}\right], \tag{A4}$$

$$b=J_0+2D+J_{OH}H_L-\frac{kT_L}{1-M_L^2}\ , \tag{A5}$$

$$c=J_{OH}-\frac{kT_LH_L}{16D^2(1-M_L^2)^2} \tag{A6}$$

$$d=J_{OT}-\frac{k}{1-M_L^2}\ , \tag{A7}$$

$$M_L=\frac{H_L}{4D}\ . \tag{A8}$$

APPENDIX B

The paramagnetic free energy and the coefficients of the Landau expansion, Eq. (3.19), are given as

$$N^{-3}G_0=-kT\ln 2+\tfrac{1}{2}kT[(1+M)\ln(1+M)+(1-M)\ln(1-M)]-\tfrac{1}{2}\hat{J}(0)M^2+DM^2-HM\ , \tag{B1}$$

$$A_{20}=\tfrac{1}{2}[\hat{J}(0)-2D+HM^{-1}]\ , \tag{B2}$$

$$A_{02}=\frac{kT}{2(1-M^2)}\ , \tag{B3}$$

$$A_{21}=\frac{1}{2M}\left[\frac{kT}{1-M^2}-\hat{J}(0)+2D-HM^{-1}\right], \tag{B4}$$

$$A_{03}=\frac{kTM}{3(1-M^2)^2}\ , \tag{B5}$$

$$A_{40}=\frac{1}{8M^2}\left[\frac{kT}{1-M^2}-\hat{J}(0)+2D-HM^{-1}\right], \tag{B6}$$

$$A_{22}=\frac{kT}{2(1-M^2)^2}-\frac{3kT}{4M^2(1-M^2)}+\frac{3[\hat{J}(0)-2D+HM^{-1}]}{4M^2}\ , \tag{B7}$$

$$A_{04}=\frac{kT}{12}\frac{1+3M^2}{(1-M^2)^3}\ , \tag{B8}$$

$$B_{10}=\frac{1}{2}\left[6A_{40}-\frac{A_{21}^2}{B(q_c)}\right]. \tag{B9}$$

APPENDIX C

The coefficients in the expression for the uniform transverse susceptibility, Eqs. (4.6) and (4.7), are

$$\mathcal{M}=\frac{3A_{40}-\dfrac{A_{21}^2}{2B(q_c)}}{6A_{40}-A_{21}^2\left[\dfrac{1}{B(0)}+\dfrac{1}{2B(2q_c)}\right]}\ , \tag{C1}$$

$$C'=2C_0\left[\mathcal{M}\frac{\partial A(q_c)}{\partial H}\right]_{T,H_\lambda(T)}. \tag{C2}$$

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