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Dietrich Belitz

Theodore R. Kirkpatrick

Thomas Vojta
Missouri University of Science and Technology, vojtat@mst.edu

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First Order Transitions and Multicritical Points in Weak Itinerant Ferromagnets

D. Belitz* and T. R. Kirkpatrick†
Institute for Theoretical Physics, University of California, Santa Barbara, California 93106

Thomas Vojta
Institut für Physik, TU Chemnitz, D-09107 Chemnitz, Germany
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It is shown that the phase transition in low-T_c clean itinerant ferromagnets is generically of first order, due to correlation effects that lead to a nonanalytic term in the free energy. A tricritical point separates the line of first order transitions from Heisenberg critical behavior at higher temperatures. Sufficiently strong quenched disorder suppresses the first order transition via the appearance of a critical end point. A semiquantitative discussion is given in terms of recent experiments on MnSi, and predictions for other experiments are made. [S0031-9007(99)09305-9]

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The thermal paramagnet-to-ferromagnet transition at the Curie temperature T_C is usually regarded as a prime example of a second order phase transition. For materials with high T_C this is well established both experimentally and theoretically. Recently there has been a considerable interest in the corresponding *quantum* phase transition of itinerant electrons at zero temperature ($T = 0$), and in the related finite T properties of weak itinerant ferromagnets, i.e., systems with a very low T_C. Experimentally, the transition in the weak ferromagnet MnSi has been tuned to different T_C by applying hydrostatic pressure [1]. Interestingly, the transition at low T was found to be of *first order*, while at higher transition temperatures it is of second order [2]. The tricritical temperature that separates the two types of transitions was found to roughly coincide with the location of a maximum in the magnetic susceptibility in the paramagnetic phase. Theoretically, it has been shown [3,4] that in a $T = 0$ itinerant electron system, soft modes that are unrelated to the critical order parameter (OP) or magnetization fluctuations couple to the latter. This leads to an effective long-range interaction between the OP fluctuations. In disordered systems, the additional soft modes are the same "diffusons" that cause the so-called weak-localization effects in paramagnetic metals [5]. In clean systems there are analogous, albeit weaker, effects that manifest themselves as corrections to Fermi liquid theory [6]. A Gaussian theory is sufficient to obtain the exact quantum critical behavior in the most interesting dimension, $d = 3$, for clean as well as for disordered systems (apart from logarithmic corrections in the clean case) [3,4].

In this Letter, we show that at sufficiently low temperatures the phase transition in itinerant ferromagnets is *generically* of first order. This surprising result is shown to be rooted in fundamental and universal many-body physics underlying the transition, viz. long-wavelength correlation effects, and, hence to be independent of the band structure. This suggests that the behavior observed in MnSi is generic, and should also be present in other weak itinerant ferromagnets. We also make detailed predictions about how quenched disorder suppresses the first order transition, which allows for decisive experimental checks of our theory.

Let us start by deriving the functional form of the free energy of a bulk itinerant ferromagnet at finite T, and in the presence of quenched disorder that we parametrize by $G = 1/\epsilon_F \tau$, where ϵ_F is the Fermi energy, and τ is the elastic mean-free time. The general Landau expansion of the free energy F as a function of the magnetic moment m in an approximation that neglects OP fluctuations is

$$F = tm^2 + u_4 m^4 + u_6 m^6 + \ldots . \tag{1a}$$

The coefficients t, u_4, u_6, etc. in this expansion can have nontrivial properties and contain important physics. A derivation from a microscopic theory shows that they are given as frequency-momentum integrals over correlation functions in a "reference system" that depends on the nature of the underlying microscopic model [7]. If the critical magnetization fluctuations are the only soft modes in the system, then they are simply numbers. However, if in the process of deriving the Landau functional some other soft modes have been integrated out, then the coefficients will, in general, not exist, since they are represented as diverging integrals over the soft modes. In Refs. [3,4] it was shown that in an itinerant electron system at $T = 0$ there are indeed such soft modes. In the disordered case, these are the diffusons mentioned above, with a dispersion relation $\omega \sim k^2$, and they lead to coefficients whose divergent parts have the form

$$u_{2m} \propto \int_0^\Lambda dk\, k^2 \int d\omega\, \frac{1}{(\omega + k^2)^{2m}} . \tag{1b}$$

Here Λ is a momentum cutoff, and all prefactors in the integrals have been omitted. In the clean case, the relevant soft modes are particle-hole excitations in the spin-triplet channel with a ballistic dispersion relation, $\omega \sim k$. The resulting integrals are still divergent, although not as strongly as in the disordered case. It was shown in Refs. [3,4] that

these divergent terms in the Landau expansion can be understood as an illegal expansion of a nonanalytic term in the free energy of the form

$$f(m) = m^4 \int_0^\Lambda dk\, k^2 \int_0^\infty d\omega\, \frac{(-1)^x}{[(\omega + k^x)^2 + m^2]^2}\,. \tag{2}$$

In the disordered case, where $x = 2$, this follows explicitly from Eq. (3.6') of Ref. [3]. In the clean case, an analogous treatment yields the same expression with $x = 1$. Notice the different sign of the dirty case compared to the clean one, which we will come back to below. Equation (2) yields $f(m) \propto m^{5/2}$ and $f(m) \propto m^4 \ln m$ in the disordered and clean cases, respectively. In either case, the resulting singularity is protected by the magnetization, which gives the soft modes a mass. The leading effect of $T \neq 0$ is adequately represented by replacing $\omega \to \omega + T$. In addition, in the presence of disorder the ballistic modes in the clean case obtain a mass proportional to $1/\tau$, so the appropriate generalization of Eq. (2) for the clean case ($x = 1$) to finite temperature and disorder is obtained by the replacement $\omega \to \omega + T + 1/\tau$. Doing the integrals, and adding the usual terms of order m^2 and m^4, we obtain a free energy of the form

$$F = tm^2 + G(N_F\Gamma_t)m^4[m^2 + (\alpha T)^2]^{-3/4} + \upsilon m^4 \ln[m^2 + (T + \beta G)^2] + um^4 + O(m^6)\,, \tag{3}$$

where Γ_t is an effective spin-triplet interaction amplitude [3] made dimensionless by means of a density of states at the Fermi level, N_F. If we measure F, m, and T in terms of a microscopic energy, e.g., ϵ_F, then t, υ, and u are all dimensionless. υ is quadratic in Γ_t [4]. $t = 1 - N_F U$ is the dimensionless distance from the critical point. It depends on the physical spin-triplet interaction amplitude U, with $N_F U \approx 1$ in a ferromagnetic or nearly ferromagnetic system, while Γ_t above is an effective interaction amplitude with $N_F\Gamma_t < 1$. Γ_t is expected to be relatively larger in strongly correlated systems. Finally, α and β are parameters that measure the relative strengths of the temperature and the disorder dependence, respectively, in the two nonanalytic terms. They are numbers of order unity, and like u and υ they are nonuniversal. Equation (3) provides a functional form of the free energy that correctly describes the leading nonanalytic m-dependence for both clean and disordered systems, as well as the leading temperature cutoff for either term and the leading disorder cutoff for the clean nonanalyticity.

The sign of υ merits some attention. Perturbation theory to second order in Γ_t yields $\upsilon > 0$ [4,8]. Further, $\upsilon > 0$ indicates a decrease of the effective Stoner coupling constant I due to correlation effects: I is a homogeneous spin susceptibility, $\upsilon > 0$ means that this susceptibility increases as the wave number increases from zero [8], and correlation effects decrease with increasing wave number. It is well known that correlation effects, in general, decrease I [9], and $\upsilon > 0$ is consistent with that. Reference [4] has given some possible mechanisms for υ to be negative at least in some materials, and shown that in this case the ferromagnetic transition is always of second order. However, the generic case is $\upsilon > 0$, which we will now discuss.

We first consider the case $T = 0$. The transition in the clean system, $G = 0$, is then of first order, since $m^4 \ln m < 0$ for small m. Upon disordering the system, $G > 0$, the negative term is no longer the leading one at $t = 0$. For small values of G, the transition remains first order. However, for G exceeding a value G_{ce} the first order transition occurs only at $t < 0$, and it is preempted by a second order transition. Since the negative term is only the third term in an m expansion of F, the multicritical point where the nature of the transition changes is a critical end point (CEP) [10]. The phase diagram in the G-t plane is shown in Fig. 1. For $G_{ce} < G < G_c$, the second order transition at $t = 0$ is followed by a second transition, the second one being of first order, to a state with a larger magnetization. The line of first order transitions ends in a critical point (CP) at a disorder value G_c, where the two minima in the free energy merge.

Before we consider $T > 0$, let us discuss this result and the validity of our conclusions. To facilitate an analytic discussion, we put $\beta = 0$. We then have $F = tm^2 + G(N_F\Gamma_t)m^{5/2} + 2\upsilon m^4 \ln m + um^4$. At $G = 0$, there is a first order transition at $t = \upsilon \exp[-(1 + u/\upsilon)]$, and the magnetization at the transition has a value $m = \exp[-(1 + u/\upsilon)/2]$. Notice that the nonanalytic term is the *leading* one in F after the tm^2 term, and that we know the functional form of F *exactly* up to $O(m^4)$. As long as $u/\upsilon \gg 1$, m is exponentially small at the transition. For small υ, our Landau expansion is therefore controlled in the sense that terms of $O(m^6)$ and higher would have to have exponentially large coefficients in order to change our results. For $G > G_{ce} = (4\upsilon/3N_F\Gamma_t)\exp[-(1 + 3u/4\upsilon)]$, the first order transition is preempted by a second order one. At the CEP, the magnetic moment has the value $m = \exp[-(2/3 + u/2\upsilon)] = e^{-1/6}\, m(G = 0)$. Allowing for $\beta = O(1) \neq 0$, and repeating the calculation numerically, leads only to minor quantitative changes of these results.

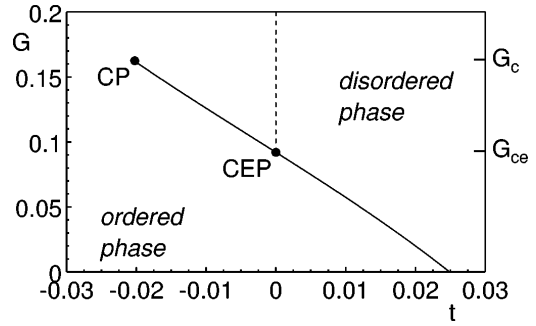

FIG. 1. Phase diagram at $T = 0$ for $u = 1$, $\upsilon = 0.5$, $N_F\Gamma_t = 0.5$, $\alpha = \beta = 1$, showing a second order transition (dashed line), and a first order transition (solid line).

At $T > 0$, the free energy is an analytic function of m, but for small T the coefficients in an m expansion become very large. Our remarks about the validity of our truncated Landau expansion therefore still apply; i.e., at $0 < k_B T \ll \epsilon_F$, our theory contains the most important terms to every order in an expansion in powers of m^2. Let us first consider the clean system, $G = 0$. There is a tricritical point (TCP) at $T_{tc} = \exp(-u/2v)$, with a first order transition for $T < T_{tc}$, and a line of Heisenberg critical points for $T > T_{tc}$. To describe the (conventional) tricritical behavior in $d = 3$ our mean-field theory is sufficient (apart from logarithmic corrections) [11]; for the critical behavior at $T > T_{tc}$ it is of course not.

For the suppression of the first order transition by disorder at $T > 0$ we find two different possibilities, depending on the value of the parameter α. For small α ($\alpha \lesssim 1.5$ with our choice of the remaining parameters, Fig. 2), the TCP is replaced by a CEP for G larger than some $G_{tce} < G_{ce}$. At $G = G_{ce}$, the CEP reaches $T = 0$, and for larger values of G the transition is of second order for all T. At small T, it is followed by a first order transition. The line of first order transitions ends in a critical point, and disappears only for $G = G_c$. For larger values of α (Fig. 3), the TCP persists for a range of disorder larger than G_{ce}. The first order transition first gets preempted in a temperature window between two CEPs. At $G = G_{ce}$, the lower CEP reaches $T = 0$, while the TCP at higher temperature survives. With further increasing disorder, two CPs appear in the ordered phase, and the remaining CEP gets replaced by a TCP. Finally, the two TCPs merge, and the remaining CP reaches $T = 0$, eliminating the last temperature regions with first order transitions. Notice that the interesting features of these phase diagrams do not depend on the logarithm in Eq. (3); similar features are obtained in standard phenomenological Landau expansions with a negative coefficient of the third term [12]. We stress again, however, that in our case the expansion is controlled, and that we have a definite physical mechanism for the appearance of a negative term, in contrast to purely phenomenological theories.

We now turn to a discussion of the available experimental information on this subject. MnSi has a low T_C ($\approx$30 K) under ambient pressure, and T_c can be driven to zero by a hydrostatic pressure $p_c \approx 15$ kbar. $k_B T/\epsilon_F \ll 1$ always, and T is low enough to suppress phase breaking processes, so the quantum critical behavior is easily accessible experimentally. This system has been studied in detail by Pfleiderer *et al.* [1] These authors found from susceptibility measurements that the transition turns first order at a T_c of about 12 K. The line of second order transitions was found to scale with pressure as $T_c \propto (p_c - p)^{3/4}$, while in the first order regime the transition temperature varies as $T_1 \propto (p - p_c)^{1/2}$. The scaling of T_c with pressure was explained by a scaling analysis based on the self-consistently renormalized (SCR) theory of Moriya and Kawabata [13], assuming a dynamical exponent $z = 3$. The first order transition at low T was attributed in Ref. [1] to a sharp structure in the density of states at the Fermi level.

Let us look at the experiment in the light of the above discussion. In Ref. [4] it was shown that the quantum phase transition in $d = 3$ is indeed correctly described by SCR theory, apart from logarithmic corrections that would be very difficult to detect experimentally, and that the dynamical critical exponent in $d = 3$ is $z = 3$. The analysis of Ref. [1] was therefore adequate, and, in particular, the quantum-to-classical crossover exponent ϕ, which determines the behavior of the critical temperature as a function of t through the relation $T_c \propto t^{\phi}$, has a value $\phi = 3/4$. If one makes the plausible assumption that t depends linearly on the hydrostatic pressure, at least for small t, then this is in agreement with both the experimental finding and the analysis in Ref. [1]. As for the pressure dependence of T_1, one of the temperature scales in the problem is the Fermi liquid temperature scale [4], which arises from a quadratic T-dependence of t. Since the first order transition is determined by the condition $t(T_1) = \text{const}$, we immediately get $T_1 \propto \sqrt{p_c - p}$, where we again assume a linear relation between p and t.

We finally discuss the observation [1] that the tricritical temperature roughly coincides with a minimum of the inverse magnetic susceptibility χ^{-1} in the paramagnetic phase. In d dimensions, the leading T-dependence of the paramagnetic susceptibility is of the form [8]

$$\chi/2N_F = 1 + 2\tilde{v}_d T^2 T^{d-3} - \tilde{u}_d T^2. \qquad (4)$$

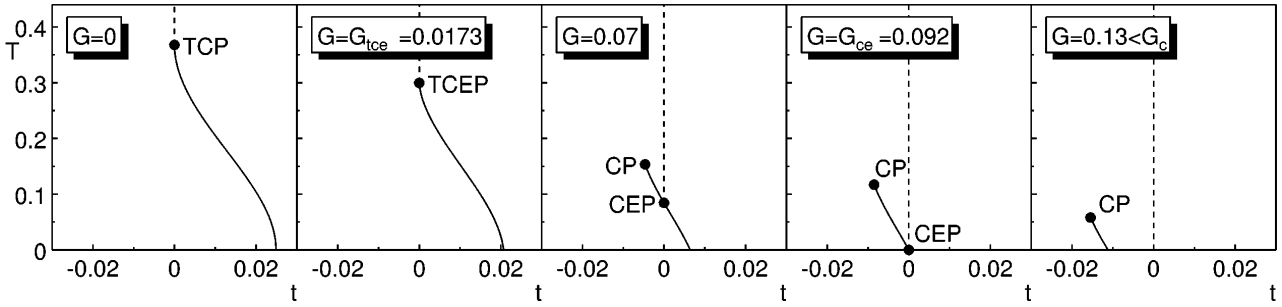

FIG. 2. Phase diagrams for $u = \beta = 1$, $v = \alpha = N_F \Gamma_t = 0.5$ showing first (solid) and second (dashed) order transitions.

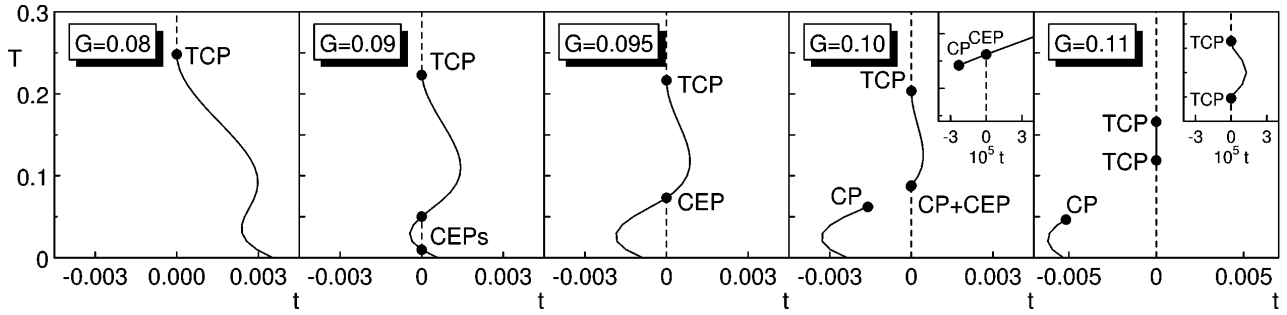

FIG. 3. Same as Fig. 2, but for $\alpha = 2$.

In $d = 3$, the nonanalyticity is of the form $T^2 \ln T$. A calculation of $\tilde{v}_3$ to second order in Γ_t revealed [8] that to that order $\tilde{v}_3 = 0$, in agreement with prior results from Fermi liquid theory [14]. Reference [8] also discussed that there are reasons to believe that the *exact* value of $\tilde{v}_3 = 0$ may be nonzero. If we assume that this is the case, then we obtain a minimum in χ^{-1} at a temperature $T_{\min} = \exp(-\tilde{u}_3/2\tilde{v}_3 - 1/2)$. Since the nonanalyticities in F and χ are manifestations of the same singularity, one expects $\tilde{u}_3 \approx u$ and $\tilde{v}_3 \approx v$, so that $T_{\min} \approx T_{\rm tc}$. While this provides a possible explanation for the observation, we stress the speculative nature of the above considerations due to the theoretical uncertainty about a nonanalytic T dependence of χ in $d = 3$.

Our theory thus provides us with a complete explanation for the nature of the transitions observed in MnSi, and, in particular, for the existence of a first order transition at low T, which in Ref. [1] was attributed to a band structure feature characteristic of MnSi. While this feature may well be sufficient to make the transition in MnSi of first order, the present theory leads to the surprising prediction that the first order transition is *generic*, and thus should be present in other weak itinerant ferromagnets as well. Our theory further predicts in detail how the first order transition will be suppressed by quenched disorder. Observations of such a suppression, or lack thereof, would be very interesting for corroborating or refuting the theory. Semiquantitatively, the theory predicts that the T region that shows a first order transition will be largest for strongly correlated systems. Conversely, since the dependence of the tricritical temperature on the system parameters is exponential, in some, or even many, systems the first order transition may take place only at very low temperatures. This may explain why in $ZrZn_2$ no first order transition has been observed [1], although the experiment does not seem to rule out a weakly first order transition [15].

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*Permanent address: Department of Physics, and Materials Science Institute, University of Oregon, Eugene, OR 97403.

†Permanent address: Institute for Physical Science and Technology, and Department of Physics, University of Maryland, College Park, MD 20742.

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[2] MnSi is actually a helimagnet, which may lead to a weakly first order transition even for higher T_C. The helimagnetism is a weak effect that is destroyed by very small amounts of disorder [1]. We will concentrate on the much stronger effects reported by these authors, and leave for future work the interesting question of if and how the helimagnetism influences the quantum transition.

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