Tricritical Behavior in Itinerant Quantum Ferromagnets

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It is shown that the peculiar features observed in the low-temperature phase diagrams of $ZrZn_2$, UGe_2, and MnSi can be understood in terms of a simple mean-field theory. The nature of the ferromagnetic transition changes from second order to first order at a tricritical point, and in a small external magnetic field surfaces of first-order transitions emerge which terminate in quantum critical points. This field dependence of the phase diagram follows directly from the existence of the tricritical point. The quantum critical behavior in a nonzero field is calculated exactly.

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The ferromagnetic transition at the Curie points of the elements Fe, Ni, and Co is one of the best-known examples of a second-order phase transition. It is well understood in terms of the band theory of metals in conjunction with the theory of phase transitions [1]. Recent experimental studies of ferromagnetic compounds with much lower Curie temperatures, among them $ZrZn_2$ [2], UGe_2 [3], and MnSi [4,5], show enigmatic behavior which does not seem to fit into this well-established picture: If the low Curie temperature is further decreased by means of pressure tuning, the nature of the transition changes from second order to first order at a tricritical point, and in a small external magnetic field h surfaces or ''wings'' of first-order transitions emerge which extend from the coexistence line at zero field and terminate in quantum critical points. These regions of first-order transitions end in lines of critical points which are reminiscent of conventional liquid-gas critical points, and which connect the tricritical point with two quantum critical points in the zero-temperature plane. These observations are summarized in the schematic phase diagram shown in Fig. 1.

This structure of the phase diagram is very remarkable for two reasons. First, the ferromagnetic transition in zero field at high-temperature Curie points, most notably in the elemental ferromagnets, is invariably of second order. Also, Hertz's theory of the quantum ferromagnetic transition at $T = 0$ [6], and its extension to nonzero temperature [7], predicts the ferromagnetic transition to be generically of second order. Second, the persistence of the first-order transition away from the zero-field plane, and the existence of quantum critical points at $h \neq 0$, came as a surprise [8]. Yet the observed structure of the phase diagram as sketched in Fig. 1 seems to be generic, as demonstrated by the case of $ZrZn_2$ as the latest example, where a tricritical point emerged once sufficiently clean samples were produced [2].

In this Letter we show that all of these observations can be explained by a theory that takes into account the fact that, in metallic systems at low temperatures, the particle-hole excitations characteristic of systems with a Fermi surface couple to the fluctuations of the magnetic order parameter and substantially change the nature of the phase transition compared to the conventional theory [9,10]. Furthermore, we identify the universality classes for all finite-temperature critical points in the phase diagram, and we determine the exact critical behavior at the quantum critical points.

Within the framework of this theory, a mean-field expression for the free energy density can be derived. In three-dimensional systems it is given by

$$f = -h\phi + t\phi^2 + v\phi^4 \ln(\phi^2/m_0^2 + T^2/T_0^2) + u\phi^4 \quad (1)$$

in terms of an order parameter ϕ. For $v = 0$ this is the usual Landau free energy [11]. t and u are coefficients that describe the distance from criticality and the importance of

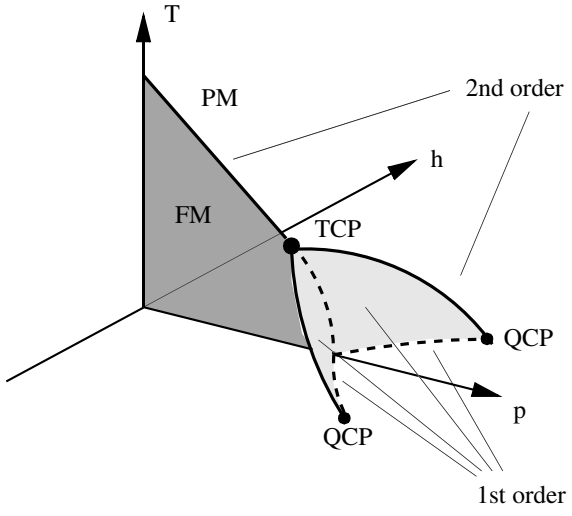

FIG. 1. Schematic phase diagram in the temperature-pressure-magnetic-field ($T - p - h$) space. Shown are the ferromagnetic (FM, dark shaded) and paramagnetic (PM) phases at $h = 0$, the tricritical point (TCP), and the two quantum critical points (QCP). Also shown are various lines of first-order (dashed lines) and second-order (solid lines) phase transitions, and the ''wing'' surfaces of first-order transitions (light shaded).

fluctuations, respectively, and h is the external field conjugate to the order parameter. For later reference we note that within Stoner theory [12,13], which is a particular realization of Landau theory, $t = 1 - \Gamma_t N_F$, with N_F the density of states at the Fermi surface and Γ_t a microscopic spin-triplet interaction amplitude, and u is proportional to the second derivative of the density of states. The parameter v reflects mode-mode coupling effects that are neglected in Landau theory. These effects are related to nonanalytic corrections to the leading behavior of correlation functions in Fermi-liquid theory [14–16]. Their precursors can also be found in the generalizations of Stoner theory that took into account spin-fluctuation effects [17]. The coupling constant v is quadratic in the microscopic coupling constant Γ_t [18]. v is thus expected to be small compared to u, and larger in strongly correlated systems than in weakly correlated ones [9]. m_0 is a microscopic magnetization (e.g., one Bohr magneton μ_B per volume of a unit cell), and T_0 is a microscopic temperature (e.g., the Fermi temperature). For $h = 0$ the free energy given by Eq. (1) was first considered in Ref. [18]. Equation (1) constitutes a natural guess for the generalization to $h \neq 0$, which is confirmed by a derivation along the same lines as in Refs. [9,10]. The physical value of ϕ is the one that minimizes f; it is equal to the magnetization m [19]. The equation of state, which relates m, T, and h, is obtained by minimizing f, which leads to

$$h = 2tm + 4vm^3 \ln(m^2/m_0^2 + T^2/T_0^2) + 2vm^3 \frac{m^2/m_0^2}{m^2/m_0^2 + T^2/T_0^2} + 4um^3. \quad (2)$$

Notice that, at $T = 0$, both the free energy and the equation of state are nonanalytic functions of the order parameter by virtue of the logarithmic term. This is in sharp contrast to ordinary Landau theory [11], where f is an analytic function of ϕ, and it reflects the fact that the particle-hole excitations have been integrated out to obtain a free energy in terms of the order parameter only. This is a particular example of a more general phenomenon; see Ref. [20].

In order to discuss the mean-field theory given by Eq. (2), we first recall the solution at $h = 0$ [18]. There is a tricritical point at $(t = 0, T = T_{tc})$, with $T_{tc} = T_0 e^{-u/2v}$. At $T = 0$, the transition occurs at $t = t_1 = m_0^2 e^{-1} v e^{-u/v}$ and is of first order with the magnetization changing discontinuously from $m = 0$ to $m = m_1 = m_0 e^{-1/2} e^{-u/2v}$. The line of first-order transitions at temperatures $T < T_{tc}$ is determined by $f(m) = f'(m) = 0$ and can be given explicitly in parametric form $(0 \leq s \leq 1)$:

$$t(s) = t_1(1 - s)^2 e^s, \quad (3a)$$

$$T(s) = T_{tc} s^{1/2} e^{-(1-s)/2}. \quad (3b)$$

We now turn to the properties of the equation of state for $h \neq 0$. Consider first the $T = 0$ plane. Suppose t has been tuned to t_1, so that at $h = 0$ the free energy has two degenerate minima, one at $m = 0$ and one at $m = m_1$. For a small $h > 0$ this double-minimum structure persists, and the two minima can still be made degenerate by increasing t; see Fig. 2. Thus there is still a first-order transition. However, with increasing h the two minima merge at a point where the first three derivatives of f vanish: $f'(m_c) = f''(m_c) = f'''(m_c) = 0$ with $m_c = m_0 e^{-13/12} e^{-u/2v}$. This condition determines a critical point (t_c, h_c) in the $T = 0$ plane that marks the end point of a line of first-order transitions. One finds

$$t_c = 6vm_c^2 = 6e^{-13/6} m_0^2 v e^{-u/v}, \quad (4a)$$

$$h_c = \frac{16}{3} v m_c^3 = \frac{16}{3} e^{-13/4} m_0^3 v e^{-3u/2v}. \quad (4b)$$

This discussion can be repeated for any fixed value of $T < T_{tc}$. Accordingly, there is a line of critical points connecting the tricritical point at $(T = T_{tc}, t = 0, h = 0)$ and the quantum critical point at $(T = 0, t = t_c, h = h_c)$. A parametric representation for this line is

$$t(s) = t_1(4s^2 + 5s + 6)(1 - s)^2 e^{1+g(s)}, \quad (5a)$$

$$T(s) = T_{tc} s^{1/2} e^{g(s)/2}, \quad (5b)$$

$$h(s) = h_c(s^2 + s + 1)(1 - s)^{5/2} e^{13/4+3g(s)/2}, \quad (5c)$$

where

$$g(s) = \frac{-1}{6}(4s^2 + 7s + 13)(1 - s). \quad (5d)$$

This line forms a boundary of a surface of first-order transitions that is shown in Fig. 3.

We see that the phase diagram obtained from Eq. (2) has the same structure as the one observed experimentally; see Fig. 1. As is the case with Landau theory, the phase diagram in a space spanned by the observables T, p, and h will be a stretched and rotated version of the one in $T - t - h$ space, since the parameters of the theory are com-

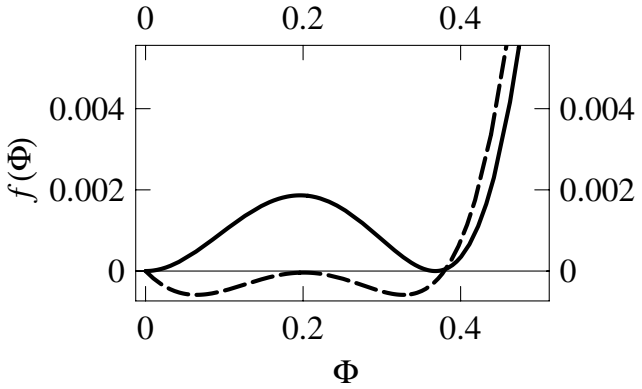

FIG. 2. The free energy at $T = 0$ for $h = 0$, $t = t_1$ (solid curve) and $h = 0.02$, $t = 0.1878$ (dashed curve), respectively. In both cases, $u = v = m_0 = 1$ and $T = 0$.

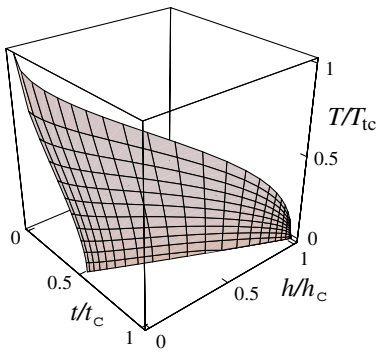

FIG. 3 (color online). The surface of first-order transitions in the space spanned by T, t, and h. It is bounded by lines of first-order transitions in the $T = 0$ and $h = 0$ planes, respectively, and by the line of second-order transitions discussed after Eqs. (4). A symmetric surface extends into the region where $h < 0$.

plicated functions of pressure, temperature, and magnetic field [21]. However, a quantitative comparison with experiment can be made by expressing the zero-temperature critical field strength h_c, Eq. (4b), in terms of observable quantities, namely, the discontinuities of the magnetization and the inverse magnetic susceptibility, respectively, across the first-order transition at $T = h = 0$. The former is given by m_1, and differentiating the equation of state shows the latter to be $\Delta\chi^{-1} = 2\upsilon m_1{}^2$. We find

$$h_c = \tfrac{8}{3}e^{-7/4}m_1\Delta\chi^{-1} \approx 0.46 m_1\Delta\chi^{-1}. \tag{6}$$

A very rough estimate using the data from Ref. [2] predicts a value of h_c on the order of 0.1 T for $ZrZn_2$.

We now turn to the critical behavior at the various critical points in the phase diagram. Equation (2) yields mean-field critical behavior for all points on the lines of critical points at $T > T_{tc}$ and $T_{tc} > T > 0$. In particular, the order parameter critical exponents β and δ have their mean-field values $\beta = 1/2$ and $\delta = 3$, respectively. At the tricritical point one finds mean-field tricritical behavior [22,23], with $\beta = 1/4$ and $\delta = 5$. This behavior gets modified if fluctuations are taken into account. For $T > T_{tc}$ the upper critical dimension d_c^+, above which mean-field critical behavior is exact, is $d_c^+ = 4$. For $d = 3$ the exact critical behavior is in the classical Heisenberg, XY, or Ising universality class, depending on the nature of the ferromagnet. ($ZrZn_2$ is a Heisenberg magnet; UGe_2 has a strong spin anisotropy and is thus Ising-like; MnSi is a weak helimagnet, which leads to some complications which we ignore here [5].) For the wing-critical lines at $T < T_{tc}$, $d_c^+ = 4$ as well. The exact critical behavior is always in the Ising universality class, since the external magnetic field reduces the effective dimension of the order parameter to one. At the tricritical point, $d_c^+ = 3$, and the mean-field theory yields the exact critical behavior except for logarithmic corrections to scaling [24].

For the quantum critical behavior at $T = 0$ the mean-field theory yields the usual mean-field values for the static exponents, e.g., $\beta = 1/2$ and $\delta = 3$. For the temperature dependence of the order parameter at the critical point one finds $\delta m(t_c, h_c, T) \propto -T^{2/3}$, where $\delta m = m - m_c$. In the light of Ref. [10] one might expect the exact critical behavior to differ strongly from these results. However, a detailed analysis shows that Hertz theory [6,7] holds at this quantum critical point. The reason is that the nonzero magnetic field and magnetization suppress the soft-mode effects which invalidate Hertz theory, and ultimately destroy the quantum critical point, at $h = 0$. More generally, it was shown in Ref. [25] that Hertz theory is valid if the field conjugate to the order parameter does not change the soft-mode structure of the system. In the present case, an expansion in powers of δm about the quantum critical point shows that the quantity $2m_c\delta t - \delta h$, with $\delta t = t - t_c$ and $\delta h = h - h_c$, plays the role of the conjugate field. Switching on an external magnetic field from $h = 0$ gives certain soft modes a mass, but changing h from $h_c \neq 0$ does not lead to further changes in the soft-mode spectrum, and neither does changing the value of t. Mean-field theory thus gives the exact static quantum critical behavior, in particular,

$$\beta = 1/2, \qquad \delta = 3. \tag{7}$$

However, the dynamic quantum critical behavior, i.e., the temperature dependence at criticality, is modified from the mean-field result [7,26], since the leading temperature dependence of the parameter t appears only at one-loop order. This fluctuation effect leads to a temperature scale with a scale dimension $[T]_{fluc} = 9/(d + 1)$. For $d < 5$ this dominates the Fermi-liquid temperature scale, which has $[T]_{FL} = 3/2$ and is responsible for the temperature dependence of the order parameter within mean-field theory. In $d = 3$ we thus have the exact result

$$\delta m(t_c, h_c, T) \propto -T^{4/9}. \tag{8}$$

Notice that the static order parameter does not depend on the critical temperature scale, which determines the dynamical critical exponent z proper,

$$z \equiv [T]_c = 3. \tag{9}$$

For $d > 2$, the critical scale dominates the fluctuation scale for all observables that depend on it, e.g., the specific heat [7,26]. Notice that the above results are the *exact* quantum critical behavior.

We finally discuss the relation between the theory presented above and a competing mean-field theory with a very different microscopic underpinning. Sandeman *et al.* [13] have proposed a Stoner model where the equation of state is analytic in the order parameter, but band-structure effects, in particular, a double-peak structure in the density of states near the Fermi level, lead to signs of the coefficients consistent with a first-order transition. These authors

have shown that this provides an explanation, not just for the first-order nature of the paramagnet-to-ferromagnet transition, but also for a second, metamagnetic, transition observed in the ferromagnetic phase of UGe_2, and they have argued that it also leads to triplet superconductivity within the ferromagnetic state, in agreement with observations on UGe_2 and URhGe. Band-structure calculations for these two materials have confirmed that a double-peak structure near the Fermi level exists [27]. It is interesting to compare various features and predictions of these two theories.

(1) The Stoner theory relies on detailed band-structure effects to explain the first-order nature of the transition. The present theory, on the other hand, is based on a universal many-body effect, namely, the existence of soft particle-hole excitations, which are *always* present in metals. It therefore predicts the first-order transition to be a *generic* feature of low-T_c itinerant ferromagnets, independent of the details of the band structure. (2) Within the Stoner theory one expects a temperature dependence of the coefficient u from Fermi-liquid theory [28], namely, $u = u_0 - u_1(T/T_0)^2$. Here T_0 is the same microscopic temperature scale as in Eq. (1), and u_0/u_1 is on the order of unity. One therefore expects T_{tc} to be generically on the order of T_0, and it is *a priori* not clear what suppresses T_{tc} to the observed values around 10 K. The many-body theory, on the other hand, provides a natural explanation for this effect: The coefficient v in Eq. (1) reflects a mode-mode coupling effect, and therefore $v/u \ll 1$ [9]. T_{tc} is thus exponentially small compared to T_0. (3) Both theories yield magnetic-field dependences of the phase diagram that are qualitatively the same and quantitatively very close to one another. For instance, the relation given by Eq. (6) is the same in the Stoner theory, only the coefficient changes to $2^{11/2}/3 \times 5^{5/2} \approx 0.27$. Notice that no magnetic-field dependence of the coefficients of either theory is necessary in order to produce the characteristic ''wing structure'' of the phase diagram, the term $-h\phi$ in the free energy suffices. In fact, the wing structure is a direct consequence of the existence of the tricritical point [22] and will be present in any theory that describes the latter [29].

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