Quantum Triple Point and Quantum Critical End Points in Metallic Magnets

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Phys. Rev. Lett. **119**, 267202 — Published 26 December 2017

DOI: 10.1103/PhysRevLett.119.267202

Quantum triple point and quantum critical endpoints in metallic magnets

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(Dated: November 29, 2017)

In low-temperature metallic magnets, ferromagnetic (FM) and antiferromagnetic (AFM) orders can exist, adjacent to one another or concurrently, in the phase diagram of a single system. We show that universal quantum effects qualitatively alter the known phase diagrams for classical magnetsi: They shrink the region of concurrent FM and AFM order, change various transitions from second to first order, and, in the presence of a magnetic field, lead to either a quantum triple point where the FM, AFM and paramagnetic phases all coexist, or to a quantum critical end point.

Quantum phases of matter, and the quantum phase transitions (QPTs) between them, are of great current interest [1]. One area that has received much attention is metallic quantum magnets, which are known for very complex phase diagrams as a function of temperature, magnetic field, and non-thermal control parameters such as pressure or chemical composition [2]. Apart from intrinsic interest in magnetism, their understanding is important for such diverse problems as high-T_c superconductivity, iron-based superconductors, and Kondo systems. The QPT from a quantum ferromagnet (FM) to a paramagnet (PM) by now is well established to be generically discontinuous or first order as a result of soft or massless excitations in zero-temperature metals that couple to the long-wavelength magnetization fluctuations. This was predicted theoretically by Belitz, Kirkpatrick and Vojta (BKV) [3, 4], and confirmed by numerous experiments [2]. This is a dramatic departure from the second-order transition observed at higher temperatures, which is qualitatively described by a classical Landau theory. The reconciliation between the two is provided by a quantum tricritical point (QTCP; see Ref. 5 for our use of the "quantum" prefix) in the T-dependent phase diagram. In an applied magnetic field h tricritical wings appear that end in a quantum wing-critical point (QWCP) [6]; this also has been observed [2].

Quantum antiferromagnets (AFMs) represent a very different physical situation. Here the order parameter (the staggered magnetization) is a short-wavelength quantity that the fermionic soft modes do not couple to directly, and the AFM-PM is generically continuous or second order. This dichotomy raises interesting questions for systems where both AFM and FM orders are present, either in adjacent phases or concurrently in the same phase [7]. Such materials range from relatively simple compounds, such as FeRh [8] and $NbFe_2$ [9], to more complex Kondo-lattice systems such as CeRuPO [10] and $CeAgSb_2$ [11]. The transition from a pure FM to a pure AFM in clean systems is usually observed to be discontinuous, although in systems that contain substantial amounts of disorder, such as Mn-doped Ni_2MnGa [12], $Ba_{0.6}K_{0.4}Mn_2As_2$ [13], and $CaRu_{1-x}Mn_xO_3$ [14], there may be a continuous transition from a pure FM phase to a phase of concurrent FM and AFM orders. The phase diagrams are complicated, have been only incompletely mapped out for most systems, and contain a complex mixture of first and second-order transitions. One of the most detailed phase diagrams is provided by a recent study of $LaCrGe_3$ under pressure and in a magnetic field, which found an AFM dome at high pressure, with an adjacent FM phase and a first-order line of metamagnetic transitions extending from the dome boundary to higher magnetic fields [15].

These observations raise important questions, including: (1) What is the generic topology of quantum phase diagrams involving both AFM and FM order? (2) What is the nature of the various QPTs? (3) Why does concurrent FM+AFM order rarely occur, so it draws considerable attention when it does [13, 16, 17]?

In this Letter we investigate these questions and discuss a free-energy functional that answers them, is in good agreement with existing experiments, and makes predictions for future ones. For three-dimensional (3-D) systems the free-energy density has the form

$$\begin{aligned} f = r\,n^2 \;&+\; t\,m^2 + \tilde{v}\,m^2(m^2+n^4)\ln(m^2+n^4+T^2) \\ &+\; u\,n^4 + v\,m^4 + 2w\,n^2m^2 - h\,m\;. \end{aligned} \tag{1}$$

Here m and n are the average magnetization and staggered magnetization, respectively, h is the external magnetic field, and T is the temperature, all measured in suitable microscopic units. The interpretation of the parameters r, t, u, and v is the same as in an ordinary Landau theory for FM or AFM order. r and t depend on T and other control parameters, such as pressure p, in complicated ways. At critical values of these parameters, $r_c(T_c, p_c)$, $t_c(T_c, p_c)$, which in turn correspond to critical values of T and p, phase transitions occur that can be second order or first order. u and v depend on the control parameters in less crucial ways; we only need to assume that they are positive (otherwise one needs to keep term of higher order in m and n). w parameterizes the free-energy cost of concurrent FM and AFM orders: Large and small values of w penalize and favor concurrent orders, respectively. Stability requires $w > -w^*(u, v, \tilde{v})$; in the classical case one has $w^*(u, v, \tilde{v} = 0) = \sqrt{uv}$. For

$\tilde{v} = 0$ this is the classical Landau theory discussed by Moriya and Usami (MU) [18, 19], which is analytic in n and m. The quantum effects that are crucial for understanding the low-temperature phase diagrams are contained in the logarithmic term with coupling constant $\tilde{v}$. For a derivation of this term, see the Supplemental Material, which includes Refs. [20–26]; here we confine ourselves to some plausibility arguments. For a pure FM, $n = 0$, Eq. (1) reduces to the theory of BKV, and the basic question is how an AFM order parameter enters this term. It cannot do so in the same way as the FM order parameter, since it is characterized by a large wave number. However, a pair of AFM order parameters can combine to couple to both the homogeneous FM order and the fermionic soft modes, which suggests that n^2 enters the logarithmic term in the same way as m does. The derivation confirms this, and yields a positive coupling constant $\tilde{v} > 0$ that measures the strength of the quantum fluctuations. The nonanalytic nature of the quantum term, which is in sharp contrast to Landau theory, reflects the fact that soft or massless excitations have been integrated out to derive it. We note that the n^4 term multiplying the logarithm is of higher order in the order parameters and should not be taken seriously. It has no qualitative effects for our discussion. Finally, we note that in 2-D systems the quantum effects are stronger, and the logarithmic term with coupling constant $\tilde{v}$ in Eq. (1) gets replaced by

$$-\tilde{v}\, m^2 (m^2 + n^4)^{1/2} \ .$$

We now compute 3-D phase diagrams by minimizing the free-energy functional f with respect to m and n. For simplicity, we will do so for $T = 0$. For fixed nonzero T the results are qualitatively the same as long as T is smaller than a threshold value related to a tricritical temperature. The behavior in the vicinity of that temperature, as well as the stronger quantum effects in 2-D, will be discussed elsewhere [27].

r-t phase diagrams: For the most basic phase diagram in the plane spanned by r and t at $h = 0$, with all other parameters fixed, there are two possibilities: i) A single discontinuous QPT from a pure FM state to a pure AFM state, or, ii) a continuous QPT from a FM phase to an FM+AFM phase, followed by a discontinuous transition to a pure AFM phase. Which of these possibilities is realized depends on the parameter w in Eq. (1). For w larger than a critical value one has the situation shown in Fig. 1(a). There is a single transition from FM to AFM, and no FM+AFM phase occurs. A qualitative change compared to the classical phase diagram discussed by MU, shown in the inset, is that the FM-PM transition is first order due to the quantum fluctuations. As a result, the bicritical point (BCP) in the classical phase diagram is replaced by a quantum critical end point (QCEP) [5, 28]. Quantitatively, the quantum fluctuations enlarge the FM phase at the expense of

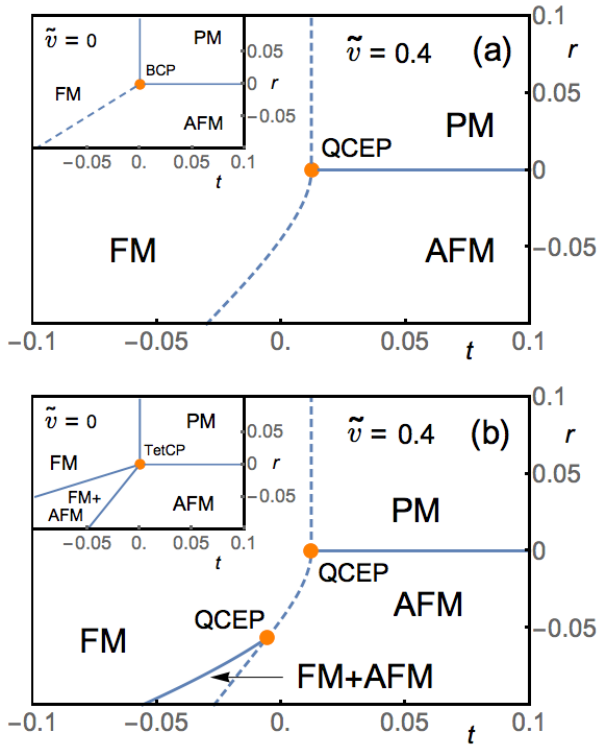

Figure 1: Phase diagrams in the r-t plane for the quantum ($\tilde{v} = 0.4$, main panels) and classical ($\tilde{v} = 0$, insets) free-energy functionals for the large-w (a) and small-w (b) case. Dashed and solid lines denote first and second-order transitions, respectively. QCEP denotes quantum critical end points, BCP denotes a bicritical point, and TetCP a tetracritical point; see Refs. 5, 28 for the nomenclature used. Parameter values are $u = v = 1$, $w = 2$ for panel (a) and $w = 0.5$ for panel (b).

the AFM one. For w smaller than the critical value one has the situation shown in Fig. 1(b), with an FM+AFM phase in between the FM and AFM phases in a part of the phase diagram. In a qualitative change from the classical phase diagram discussed by MU, which is shown in the inset, a direct FM-to-AFM transition exists, there are two QCEPs instead of a single tetracritical point (TetCP), and the existence of this phase is restricted to sufficiently negative values of r. The latter feature can be understood from a basic property of the free energy: Classically, for any solution with $m > 0$ and $n > 0$ one has $n^2 = (-r - 2wm^2)/2u$, and a relation of the same structure remains true in the quantum case. The quantum fluctuations make m discontinuous, which implies that n can be real, and the FM+AFM solution can exist, only for sufficiently large negative r. In addition to the FM-to-PM transition, those from FM to AFM, and from FM+AFM to AFM, are all first order as a result of the quantum fluctuations; the latter thus drastically change the nature of the phase diagram. We note that across the first-order FM-AFM transition in Fig. 1(b) the AFM order parameter is discontinuous, just as the FM one is.

This is an example of quantum fluctuations driving an AFM transition first order even though they couple only indirectly to the AFM order parameter.

In order to discuss phase diagrams directly relevant to experiments, consider a control parameter p on which both t and r depend. Changing p at fixed low T will thus map out a path in the t-r plane. In an actual experiment, p is often, but not necessarily, realized by hydrostatic pressure [2]. For simplicity, consider linear paths:

$$r(p) = r_0 + (r_1 - r_0)p \quad , \quad t(p) = t_0 + (t_1 - t_0)p \; . \quad (2)$$

h-p phase diagrams: In the presence of a magnetic field h the free-energy landscape contains a metamagnetic first-order transition, i.e., a discontinuity in the FM order parameter, that corresponds to the tricritical wing in the purely FM theory. This transition may or may not be physically realized, depending on whether or not the global minimum of the free energy corresponds to AFM order. For relatively large w, there are three possibilities: i) For small $\tilde{v}$ the QWCP, which marks the end point of the tricritical wing, lies inside the AFM dome, see Fig. 2(a). In this case the metamagnetic transition including the QWCP, is not observable, and the structure of the phase diagram is qualitatively the same as in the classical MU theory [18]: The AFM dome is delineated on the left by a first-order transition to a (field-polarized) FM state, and on the right by a second-order transition to a field-polarized PM state, with a tricritical point (TCP) separating the two parts of the dome boundary. The TCP may lie to the left or to the right of the dome maximum, depending on parameters, see also Fig. 3(a) and the related discussion. ii) For larger values of $\tilde{v}$ the QWCP lies outside the AFM dome. If the tricritical wing crosses the dome boundary where the AFM becomes unstable via a first-order transition, there is a quantum triple point (QTP) where the field-polarized FM and PM phases coexist with each other and with the AFM phase. The tricritical wing now has a part that is outside of the AFM dome and hence observable, and the dome boundary consists of three parts: A first-order AFM-FM transition, a first-order AFM-PM transition, and a second-order AFM-PM transition, with the TCP that also exists in the classical phase diagram separating the latter two. This case is illustrated in Fig. 2(b). iii) For even larger values of $\tilde{v}$ the tricritical wing intersects the AFM dome in its second-order section. The dome boundary now consists of only two sections, one first order and one second order, that are separated by a QCEP, see Fig. 2(c). In all three cases, the near-linear shape of the left side of the AFM dome reflects the unobservable part of the tricritical wing inside the dome and thus is a direct consequence of the quantum fluctuations. It is in sharp contrast to the much more symmetric and evenly curved phase diagram in MU theory. We note in passing that the QWCP and the asymptotic behavior of the AFM-PM phase boundary near $p = 1$ can be determined

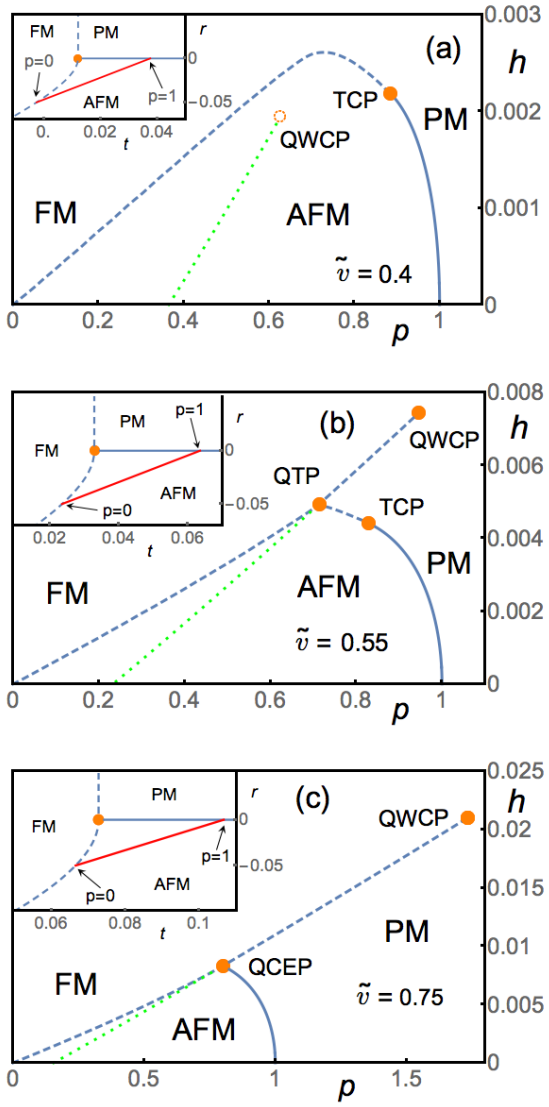

Figure 2: Phase diagrams in the h-p plane for $u = v = 1$, $w = 2$, and three different values of the quantum fluctuation parameter $\tilde{v}$. For $\tilde{v} = 0.5$, panel (a), the structure is qualitatively the same as in the classical MU theory; for larger values of $\tilde{v}$ it is drastically different. Dashed and solid lines denote first and second-order transitions, respectively. The dotted (green) line is the unobservable part of the tricritical wing inside the AFM dome and does not represent a phase transition. Special points are a tricritical point (TCP), a quantum wing critical point (QWCP), a quantum triple point (QTP), and a quantum critical end point (QCEP), see see Refs. 5, 28 for the nomenclature used. p parameterizes the linear paths in the r-t plane, Eq. (2), shown in the insets.

analytically; the other parts of the phase diagram were obtained by numerically minimizing the free energy.

For relatively small w, there are two possibilities: i) If the path in the r-t plane does not cross the FM+AFM phase, then the p-h phase diagram is qualitatively the same as in the large-w case, see Fig. 3(a), which has the same structure as Fig. 2(a). ii) If the path does cross

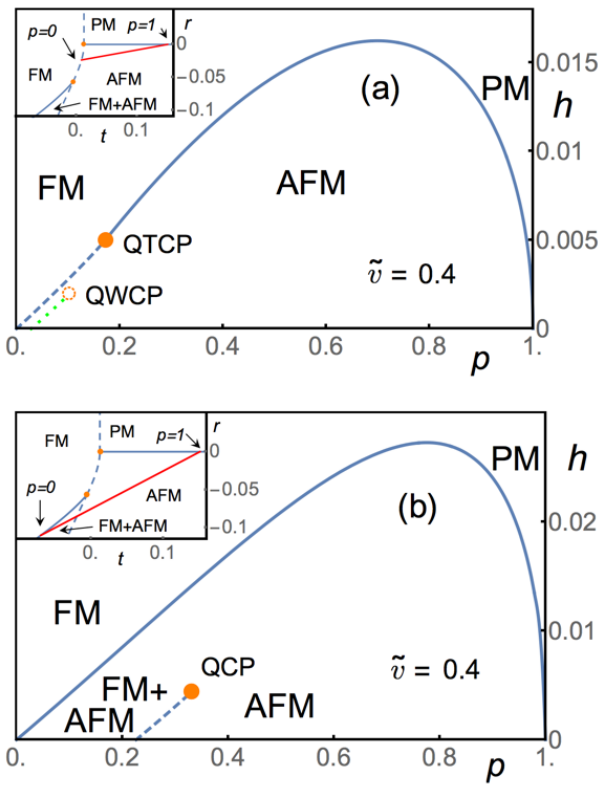

Figure 3: Phase diagram in the h-p plane for $u = v = 1$, $w = 0.5$, $\tilde{v} = 0.4$. The paths parameterized by p are shown in the insets. Solid and dashed lines denote continuous and first-order transition, respectively. QTCP and QCP denote quantum tricritical and quantum critical points, respectively, see see Ref. 5 for the nomenclature used. The dotted (green) line in panel (a) is the unobservable tricritical wing, and QWCP is the unobservable quantum wing-critical point. Note the qualitative difference between the two paths.

the FM+AFM phase, a qualitatively new feature arises: For small external fields, there is a first-order transition from the FM+AFM phase to the FM phase. This is a true phase transition within the AFM dome that has no analog in the large-w case. This line of first-order transitions ends in a quantum critical point (QCP), which in Fig. 3(b) lies within the AFM dome. This is still true for the larger values of $\tilde{v}$ used in Figs. 2(b, c). The reason is that with increasing $\tilde{v}$ the FM+AFM phase in the r-t plane is pushed to larger negative r values. The resulting increase in the h-scale that characterizes the height of the AFM dome mostly compensates for the increased size of the tricritical wing, and the first-order transition remains within the dome even for $\tilde{v} = 0.75$. With decreasing $\tilde{v}$ the length of the first-order line decreases, and in the classical case it shrinks to zero and the only transition within the dome is a critical point at $h = 0$ [18].

Relation to Experiments: Our phase diagrams are directly applicable to experimental results in T = const. planes at low T, and they are in excellent qualitative agreement with existing data. In particular, a recent experimental study of $LaCrGe_3$ has found an h-p phase diagram (p being hydrostatic pressure) consistent with Fig. 2(c), with a QWCP well outside the AFM dome [15]. For CeRuPO a phase diagram in T-p-h space has been partially mapped out [10]. In the h-p plane a metamagnetic transition was found outside the AFM dome that also is consistent with the existence of the FM-PM first order transition line in Figs. 2(b,c). Additional experimental information is desirable to determine whether the QTP case of Fig. 2(b), or the QCEP case of Fig. 2(c), is realized in this material. Experiments on other classes of materials are needed to check for phase diagrams of the type shown in Fig. 3. We note that the quantum effects are necessary to understand the observed phase diagrams: The only feature that can be understood already within the classical MU theory is the first-order nature of the FM-AFM transition, see Fig. 1(a).

Our results help explain why the FM+AFM phase is rarely seen [13, 16, 17]: It requires a special range of w-values and special properties of the path in the r-t plane, since the quantum effects push the FM+AFM phase to negative r-values of, see Figs. 1(b), 3.

As mentioned above, for temperatures that are not small compared to the tricritical temperature in the pure FM problem, qualitatively new features appear in the phase diagram including new QTCPs or QCEPs. This will be discussed elsewhere [27].

This work was supported by the National Science Foundation under Grant No. DMR-1401449 and Grant No. DMR-1401410. Part of this work was performed at the Telluride Science Research Center (TSRC). We thank Valentin Taufour for discussions.

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