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Why first order quantum phase transitions are interesting

C Pfleiderer[1]

Physikalisches Institut, Universität Karlsruhe, D-76128 Karlsruhe, Germany

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Abstract

It is frequently argued that only second order phase transitions at $T = 0$ deserve to be called quantum phase transitions, while first order quantum phase transitions are a contradiction in terms. However, quantum phase transitions differ from classical phase transitions in two fundamental ways. First, the free energy landscape need not be that of a classical second order phase transition for quantum fluctuations to drive the transition. Second, at $T = 0$ a rich variety of quantum correlation effects, such as magnetic rotons, instantons or skyrmion textures, are possible. The recent discovery of partial magnetic order, an extended non-Fermi liquid phase and superconductivity at the first order quantum phase transitions of itinerant-electron magnets underscore the need for detailed experimental studies of hitherto unexplored weak rigidities that are well known to generate first order behaviour. These include changes of the electronic valence, spin–orbit coupling, crystal electric field levels, and crystallographic structure driven by instabilities of the Fermi surface.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Over the past two decades the study of quantum phase transitions (QPT), defined as phase transitions that are driven by *quantum* fluctuations, has become a very active field of experimental and theoretical condensed matter physics [1–4a]. On the one hand, the study of quantum phase transitions has led to the discovery of novel electronic ground states in magnetic metals such as magnetically mediated superconductivity, partial or quadrupolar order and non-Fermi liquid phases. On the other hand, studies of finite temperature properties near quantum phase transitions has led to new concepts of low lying excitation spectra in condensed matter systems [4b].

[1] Present address: Physik Department E21, Technische Universität München, D-85748 Garching, Germany.

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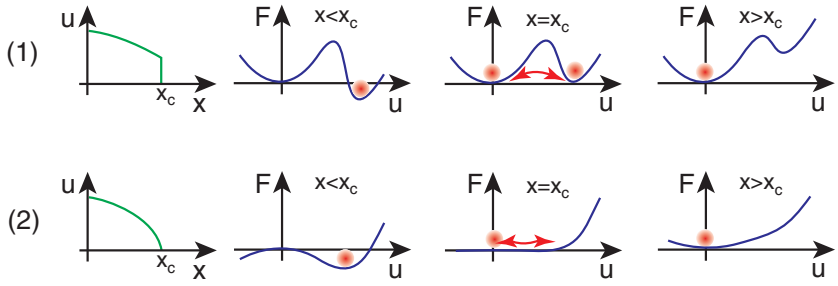

Figure 1. Qualitative illustration of the first (1) and second (2) order behaviour of the order parameter and the free energy, respectively.

It is frequently argued that the free energy landscape of quantum phase transitions has to be akin that of classical critical phase transitions to be driven by quantum fluctuations. To illustrate this issue the free energy landscape of first and second order phase transitions is illustrated in figure 1. We assume that the phase transition is characterized by the emergence of an order parameter **u** below some critical value x_c of a control parameter x. For classical phase transitions the control parameter is temperature T, in contrast to a non-thermal quantity like pressure p or uniaxial stress σ for QPT. Below x_c the order parameter appears discontinuously for first order and continuously for second order phase transitions, respectively. The corresponding free energy landscape is characterized by local minima for first order transitions while second order transitions develop a global minimum continuously. In short, classical first order transitions are driven by local minima of the free energy while classical second order phase transitions are driven by fluctuations.

In the case of a second order phase transition the order parameter fluctuations may be characterized by a relaxation frequency spectrum $\Gamma \propto q^z$, where the momentum, q, dependence defines the dynamical exponent, z. The fluctuations are referred to as 'quantum' when they are faster than the temperature of the system, $\hbar\Gamma > k_B T$. When approaching the transition temperature the relaxation frequency vanishes, $\Gamma \to 0$ for $T \to T_c$. This so-called 'critical slowing down' implies that the fluctuations ultimately change from quantum to classical for finite temperature second order phase transitions.

It was noticed by Hertz and Millis [5] that for phase transitions at $T = 0$ as a function of some non-thermal control parameter critical slowing down does no longer imply the quantum to classical crossover of the relaxation frequency spectrum. Instead the effects of quantum mechanics were shown to lead to an intimate connection of spatial and temporal order parameter variations, that may be viewed as increasing the dimension of the system by the dynamical exponent, $d_{\rm eff} = d + z$. This constitutes the conceptual advantage of quantum critical phenomena. In the majority of cases the effective dimension is *above* the upper critical dimension, $d_{\rm eff} > 4$. The fluctuations may then be accounted for in a self-consistent mean field approximation, which in turn may be equivalent to using self-consistent *linear* response theory [6].

In general, peaks of the density of states which are to be expected near the Fermi level in all real materials result in a ragged free energy landscape. While minima of the free energy may be thermally averaged out at high temperatures, they impose a trend to first order behaviour at low temperatures. However, for the case of quantum phase transitions the order parameter may explore all possible states. In particular, the system may fluctuate between states that are separated by a potential barrier. This makes first order quantum phase transitions interesting for at least three reasons. First, the characteristic relaxation frequency spectrum of fluctuations

may be completely different than hitherto assumed. Second, the nature of quantum coherence near the quantum phase transition may be completely different to anything so far assumed. Third, first order behaviour may itself be a manifestation of novel types of subtle quantum correlation effects. When taken together, this makes the physics of first order quantum phase transitions an extremely rich challenge in both theoretical and experimental studies. In the following the interest in first order QPT is illustrated by reviewing recent experimental findings in the study of itinerant-electron magnets.

2. A new generation of high pressure experiments

The recent development of a new generation of non-magnetic miniature clamp type pressure cells permitted the combination of DC magnetization and neutron diffraction experiments. The set-up was first tested in studies of the superconducting band-ferromagnet UGe_2 (D23-CRG, ILL Grenoble) [7]. Further studies have been carried out on MnSi at the cold neutron triple-axis spectrometers (4F1, LLB Saclay; TASP, PSI Villigen) [8, 9] and a small angle neutron spectrometer using polarization analysis (V4 at HMI Berlin) [10]. Recent work extended the philosophy of this work to the study of $ZrZn_2$ using a thermal triple-axis spectrometer (IN20, ILL Grenoble) [11]. The absorption by the Cu:Be clamp cell was thereby in general outweighed by the efficiency of using a well prepared experimental set-up, notably the possibility to adjust the pressure prior to the experiment and the possibility to compare the neutron intensity with the uniform magnetization.

3. Partial order in the non-Fermi liquid phase of MnSi

The transition metal compound MnSi above a certain critical pressure ($p_c = 14.6$ kbar) provides what may be the cleanest example of an enigmatic quantum phase. For these pressures the properties of MnSi challenge our understanding of metallic magnetism in two ways. First, the electrical resistivity changes abruptly from the quadratic temperature dependence characteristic of a Fermi liquid to a $T^{3/2}$ non-Fermi liquid form, providing an example of an extended non-Fermi liquid *phase* in a three-dimensional metal [12, 13]. Second, neutron diffraction measurements of MnSi reveal that sizeable quasi-static magnetic moments survive far into the non-Fermi liquid phase [8]. These moments are organized in an unusual pattern with partial order. The connection of both phenomena, likely to be carried by the same conduction electrons, is an unresolved topic of great general interest. In the following the salient features of the resistivity and partial order are reviewed to bring out aspects that are generally ignored in the study of quantum phase transitions in intermetallic compounds, notably material specific weak energy scales and weak rigidities of the long-range order underlying the quantum phase transition.

A large body of thermodynamic and microscopic data [14–23] led to the view that the ground state of MnSi below $T_c = 29.5$ K may be described as a three-dimensional weakly spin-polarized Fermi liquid *par excellence*, with the possible exception of recent high frequency optical conductivity experiments [24]. As a congruently melting compound MnSi can be produced at high purity and high crystalline perfection in the cubic B20 structure.

As one of the most important criteria that makes studies of MnSi a tractable problem, three well-separated energy and length scales can be distinguished as follows. First, MnSi has a strong tendency to itinerant ferromagnetism on length-scales of a few lattice constants, $a = 4.56$ Å, with an ordered moment of about $0.4\,\mu_B$/fu. Second, as the B20 structure lacks an inversion symmetry, weak spin–orbit interactions assume a Dzyaloshinsky–Moriya (DM) form $\int \mathbf{S}\cdot(\nabla\times\mathbf{S})\,\mathrm{d}\mathbf{r}$ which (being linear in momentum) destabilizes the uniform ferromagnetic order

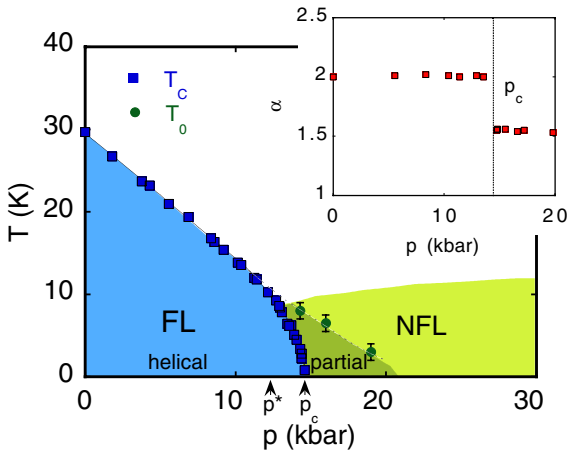

Figure 2. Phase diagram of MnSi at ambient field [8, 12, 13]. The transition temperature T_c as detected by anomalies in the resistivity and susceptibility decreases monotonically and vanishes at $p_c = 14.6$ kbar. The exponent describing the temperature dependence of the electrical resistivity, α, changes abruptly from the value of a Fermi liquid to a non-Fermi liquid form of $\alpha \approx 3/2$ when going from below to above p_c. The non-Fermi liquid phase extends at least up to $2p_c$ [30], the highest pressure studied to date, and down to the lowest temperatures of the order 20 mK.

and introduces a well-understood [25, 26] long-wavelength helical modulation of wavelength 170 Å at ambient pressure. Third, in the ordered phase, further spin–orbit interactions induced by the cubic crystalline electric fields lock the direction of the spiral to $\mathbf{Q} = \langle 111 \rangle$, where $\mathbf{S} \perp \mathbf{Q}$ [22, 23, 25, 26]. Typical sizes of magnetic domains in the ordered state are 10^4 Å [23]. The locking of the direction of the helix hence represents the weakest scale.

High sensitivity to hydrostatic pressures early suggested MnSi as a possible candidate material for the study of quantum critical phenomena [27] that are related to itinerant-electron ferromagnetism when the ordered magnetic moment disappears. In other words, it was assumed that the ordered moment vanished together with the magnetic ordering temperature T_c in strict analogy with the ordering process as a function of temperature at ambient pressure. First measurements suggested a temperature dependence of the electrical resistivity above ∼1 K consistent with the predictions of a *marginal* Fermi liquid [28] expected for this case. However detailed AC susceptibility studies in high purity single crystals soon established a change from second to first order behaviour when T_c dropped below ∼12 K [29].

Shown in figure 2 is the temperature versus pressure phase diagram of MnSi for ambient magnetic field. With increasing pressure the magnetic ordering temperature as determined from anomalies in the electrical resistivity and AC susceptibility decreases monotonically and vanishes above $p_c = 14.6$ kbar. An unexpected discovery shown in the inset was the abrupt change of the temperature dependence of the electrical resistivity in MnSi, $\Delta\rho \propto T^\alpha$ at p_c from a Fermi liquid form ($\alpha = 2$) to a stable non-Fermi liquid form ($\alpha = 3/2$) [12, 13]. The first order transition suggested the absence of scattering of the charge carriers by soft spin fluctuations expected for a collapse of the ordered moment. As a function of temperature the non-Fermi liquid resistivity has been observed down to a few millikelvins, the lowest temperatures investigated. Recent studies further establish that the non-Fermi liquid resistivity prevails up to at least 30 kbar, i.e., twice the critical pressure [30].

It has long been established that non-Fermi liquid behaviour due to an abundance of soft spin fluctuations may be readily suppressed in applied magnetic fields. This raises the

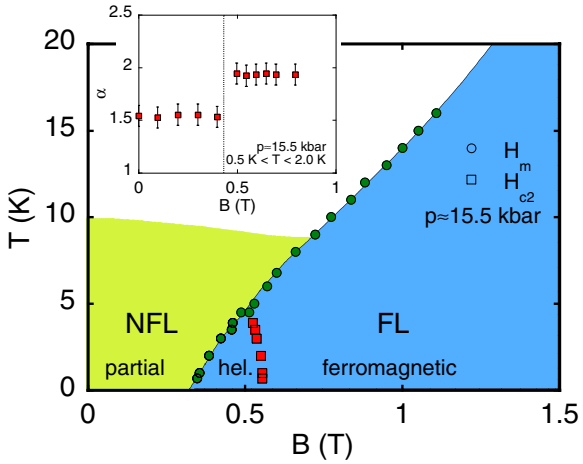

Figure 3. Magnetic phase diagram of MnSi about 1 kbar above the critical pressure for a pressure $p = 15.50$ kbar [12, 15]. The non-Fermi liquid regime (NFL) is sharply bounded by the itinerant-electron metamagnetic transition as first shown in [12]. Small angle neutron scattering shows that the magnetic state changes from partially ordered to long-range helical order at the metamagnetic transition and eventually becomes ferromagnetically spin aligned above H_{c2} as originally inferred from the AC susceptibility. The inset displays the field dependence of the exponent α, as reported in [30].

question for the evolution of the magnetic phase diagram of MnSi as a function of pressure. At ambient pressure a magnetic field induces a collapse of the helical order above a critical field $H_{c2} \approx 0.55$ T, where H_{c2} may be understood as a measure of the Dzyaloshinsky–Moriya interaction. The magnetic phase diagram of MnSi as a function of pressure was first established using high resolution AC susceptibility measurements [15]. The critical field H_{c2} was found to be unchanged as a function of pressure up to ∼16 kbar, the highest pressure studied. The change of the magnetic transition from second to first order behaviour for $p > p^* \approx 12$ kbar is accompanied by the emergence of itinerant-electron metamagnetism for $T > T_c$. Based on the AC susceptibility it has further been argued that reentrant long-range helical order exists, if the metamagnetic transition field H_m is less than H_{c2}.

The magnetoresistance for $p > p_c$ displays a pronounced drop at the metamagnetic transition field H_m. Plotting the normalized magnetoresistance for various temperatures it was concluded that an abrupt change from the non-Fermi liquid resistivity ($\Delta\rho \propto T^{3/2}$) also takes place at H_m [12]. This was confirmed in further measurements as a function of temperature for finite fields (inset of figure 3) [30]. If the non-Fermi liquid behaviour is related to a partial loss of long-range order, notably a disordering transition of the helical propagation direction, this should also be seen in neutron diffraction studies of the magnetic phase diagram.

Recent small angle neutron diffraction experiments of the magnetic field dependence confirm the prediction of reentrant helical order at H_m and provide further support for a connection of the partial magnetic order with the non-Fermi liquid phase [10]. When applying a magnetic field to the zero-field cooled state, long-range helical order is observed for magnetic fields in the range $H_m < H < H_{c2}$. On the other hand, reducing the magnetic field from above H_{c2} to zero leaves behind helical order, which disappears upon a small temperature increase. This may reflect on the creation of metastable droplets in the first order free energy landscape near p_c.

So far the focus has been the abrupt transition from a Fermi liquid to non-Fermi liquid resistivity. However, neutron diffraction studies also raise an important question about the

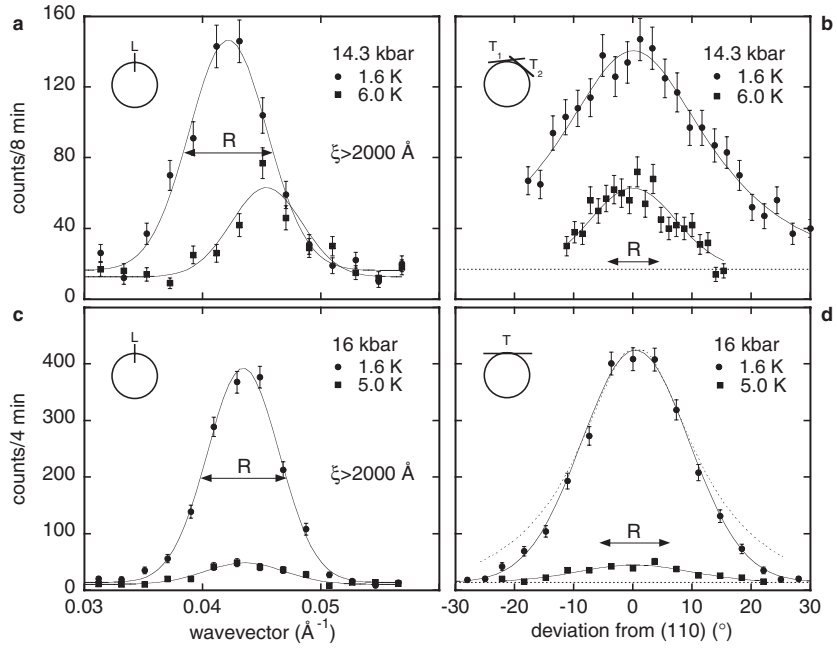

Figure 4. Neutron intensity just below and just above the critical pressure p_c, for which an abrupt change from a Fermi liquid to a non-Fermi liquid resistivity is observed [8]. Shown are typical transverse (T) and longitudinal (L) scans as depicted in the corner of each panel. The partial magnetic order emerges gradually with increasing pressure. Provided the assumption holds that the magnetic order and the electrical resistivity are carried by the same conduction electrons, this suggests that a droplet model or glassy state may not be suitable to explain the abrupt appearance of the non-Fermi liquid phase when going from below to above p_c.

Fermi liquid state below p_c. While the transition from Fermi liquid to non-Fermi liquid behaviour is abrupt, the change from long-range to partial order is gradual—perhaps even continuous. This may be illustrated by figure 4, where for $p = 14.3$ kbar just below p_c in excess of 90% of the intensity is already partially ordered. At ambient pressure the enhancement of the resistivity coefficient A is well accounted for by the abundance of soft spin fluctuations that also enhance the specific heat coefficient γ. With increasing pressure the coefficient A diverges (see figure 6 in [14]), while the ordered magnetic moment remains essentially unchanged. This suggests that the enhancement of A with pressure is not dominated by a softening of the ordered moment, but may be related to a coexistence of the long-range order with the partially ordered state. The enhancement of A may then be due to a percolation effect, where Fermi liquid behaviour is triggered by tiny volume fractions of three-dimensional long-range order. This would constitute a new mechanism causing an enhancement of Fermi liquids. It is interesting to speculate if this type of behaviour is more generally present in other intermetallic compounds, like the class of heavy-fermion systems.

An important experimental question concerns the nature of the stability of the non-Fermi liquid behaviour. We originally pointed out that the $T^{3/2}$ resistivity qualitatively matches the behaviour of spin glasses [12]. Along this line it has been suggested that the first order free energy landscape may permit the formation of metastable droplets [30]. However, droplets of three-dimensional long-range order causing the non-Fermi liquid behaviour qualitatively contradict the gradual emergence of the partial magnetic order. Further, even strongly modified

fluctuation spectra in a first order free energy landscape would be extremely sensitive to large changes of pressure, making it difficult to explain the wide range of stability of the non-Fermi liquid phase. This suggests that the partially ordered state represents a ground or lowest energy state and not just a glassy or metastable state.

The challenges posed by the phase diagram of MnSi have motivated various theoretical proposals. Rosch *et al* have considered the scattering of charge carriers by slow anomalous Goldstone modes [31] of the direction of the helix and the effects of weak spin–orbit coupling on the electronic structure [32]. Bogdanov *et al* [33] have considered the emergence of a two-dimensional magnetic structure (skyrmion textures) which are driven by the competition between ferromagnetic exchange and Dzyaloshinsky–Moriya spin–orbit coupling. Turlakov and Schmalian have considered the role of magnetic rotons for the quantum phase transition [34]. Belitz *et al*, finally, have considered non-analytic corrections to the magnetic equation of state in ferromagnets due to coupling of the magnetization to soft particle–hole excitations [35]. The latter effects may be relevant in MnSi since the helical modulation wavevector remains tiny at p_c.

For the study of quantum phase transitions in intermetallic compounds the properties of MnSi support the existence of novel metallic phases with partial ordering of the conduction electrons (reminiscent of liquid crystals), as proposed for the high temperature superconductors and heavy-fermion compounds. More generally, the interpretation of anomalous properties of intermetallic compounds to date largely ignores material specific energy scales and the particular rigidities of the ordered states that drive the quantum phase transitions. It is likely that features of the quantum phase that are stabilized in MnSi at high pressure are present in other materials, where the energy scales are, however, not well separated and therefore the partial loss of long-range order is more difficult to identify.

The role of well-separated energy scales contrasts in particular materials with equivalent energy scales like for many rare-earth heavy fermion systems or the cuprates. A challenge remains, if novel physical properties like high-T_c superconductivity may here be driven by the existence of multiple energy scales. The study of the interplay of energy scales may provide a different route to novel phases of magnetic metals.

4. Superconducting itinerant ferromagnets

Perhaps the earliest prediction of what may be called a 'novel' phase of magnetic metals near a QPT has been magnetically mediated superconductivity. The discovery of superfluidity in ^{3}He led Leggett [36] and Fay and Layzer [37] to suggest that an analogous state might exist in exchange enhanced paramagnets. The suggestion was followed up by Fay and Appel [38], who predicted that superconductivity might also exist in the ferromagnetic state. Based on a paramagnon spectrum of fluctuations described by RPA they proposed that the superconductivity in the C15 Laves phase compounds $ZrZn_2$ may be as high as 1 K *even in the ferromagnetic state*. The qualitative phase diagram proposed by Fay and Appel assumed a divergence of the longitudinal susceptibility at a ferromagnetic quantum critical point.

The theoretical proposition by Fay and Appel motivated considerable experimental efforts in the 1980s. Lonzarich and co-workers revisited the question of quantitative estimates of magnetically mediated superconductivity following the successful derivation of a quantitative model of the magnetic equation of state of weakly and nearly ferromagnetic itinerant electron magnets. To overcome the problems of low sample quality in $ZrZn_2$ together with the intense interest in heavy-fermion behaviour of f-electron materials the attention turned to lanthanide and actinide materials.

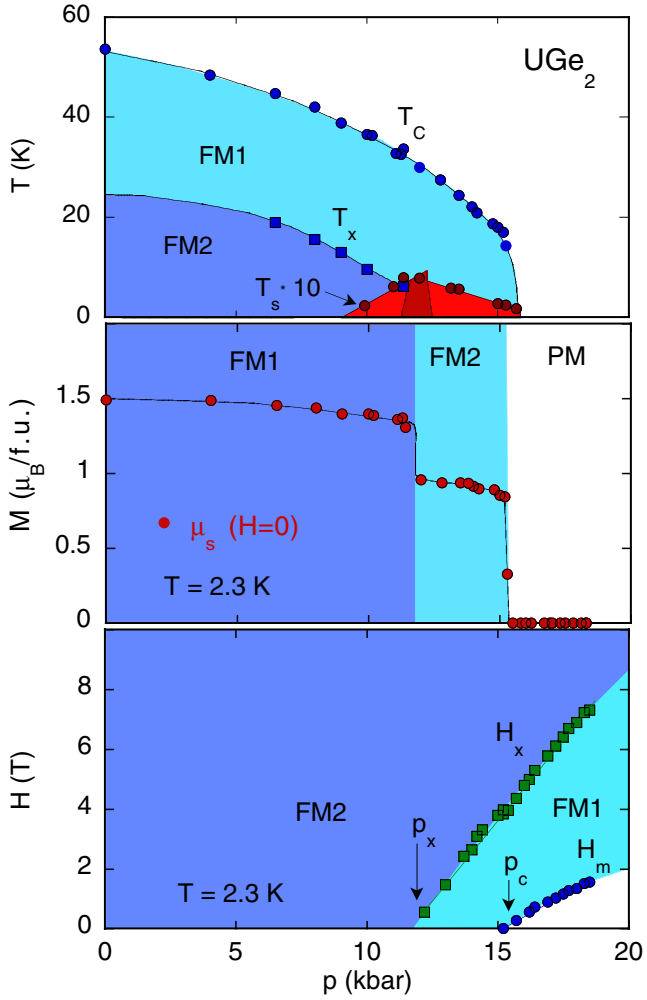

Figure 5. Phase diagram of UGe_2 as reported in [43]. The ordered magnetic moment drops discontinuously at p_x and p_c, defining two strong first order QPT. For further details refer to the text.

The discovery of superconductivity at the border of antiferromagnetism in $CePd_2Si_2$ [39] and $CeIn_3$ [40] motivated a revisit of ferromagnets, in particular the f-electron ferromagnet UGe_2 for the sake of its favourable metallurgical properties. In 1999 Argarwal and Huxley discovered superconductivity simultaneously in Cambridge and Grenoble, respectively [41, 42].

Shown in figure 5 is the phase diagram of UGe_2 as reported in [43]. Figure 5(a) shows the pressure dependence of the Curie temperature T_C and superconducting transition temperature T_s. The monotonic decrease of T_C with pressure was originally taken as evidence for a quantum critical point, where evidence for itinerant metamagnetism had hinted at a discontinuous suppression of the ordered moment [44]. First neutron diffraction studies further suggested the existence of an additional magnetic transition at temperature T_x, where signatures of T_x in the resistivity and thermal expansion may be traced all the way to ambient pressure. High pressure magnetization studies established the transition at T_x to be a line of first order transitions near p_x,

where $T_x \to 0$ [43]. Careful comparison of the (040), (100) and (111) Bragg intensities with the uniform magnetization clearly established that the two ferromagnetic phases are strongly uniaxial [7]. When taken together with measurements of the easy-axis magnetization, the FM1 phase is stronger uniaxial than the FM2 phase and has an increased orbital moment by roughly 4%.

Shown in figure 5(b) is the uniform magnetization of UGe_2 for $H \to 0$ as a function of pressure, which undergoes two distinct first order transitions at p_x and p_c, respectively. Figure 5(c) shows the metamagnetic transition fields when driving the magnetic state back to high spin-polarization with magnetic field. In the low temperature limit the metamagnetic transitions are first order over the range of pressures and fields studied here. These experiments established beyond doubt the existence of two strongly first order quantum phase transitions in UGe_2; there are no quantum critical points driving superconductivity in UGe_2.

The most perplexing observation at first appeared to be the existence of superconductivity on the ferromagnetic side of the pressure dependent phase diagram. The emergence of superconductivity in the ferromagnetic state in the presence of strong first order quantum phase transitions may be reconciled in a model proposed by Sandeman *et al* [45]. However, in the model proposed by Sandeman *et al* only the effects of spin are considered. While the model supplies a qualitative explanation it does not take into account further important material specific aspects, notably spin–orbit coupling and the presence of crystal electric fields. Further aspects of importance for UGe_2 concern the origin of the large longitudinal susceptibility and magnetic anisotropy. First, the strong uniaxial (Ising) anisotropy in UGe_2 increases slightly with pressure. This raises the question for the nature of the 'melting' of long-range order and characteristics of the magnetic domain structure at p_x and p_c, respectively. Second, the strong uniaxial character together with neutron diffraction and electronic structure calculations establishes the presence of strong spin–orbit coupling. This raises the question of the nature of the amplitude fluctuations. Third, the effects of crystal field excitations in uranium compounds which are well known to hybridize strongly with the conduction electrons and phonons are not understood.

Further uranium compounds that display superconductivity in the ferromagnetic state are URhGe [46] and UIr [47]. For URhGe high pressure studies suggest the existence of a quantum phase transition at a high extrapolated negative pressure [48]. Ferromagnetic UrIr, on the other hand, attracts interest, because superconductivity here arises in the ferromagnetic state of a material lacking space inversion symmetry. The lack of space inversion has triggered speculations of an unconventional composite superconducting order parameter.

Finally, incipient superconductivity has also been reported for high purity single crystals of $ZrZn_2$ [49, 50]. The same single crystals also showed quantum oscillations [51], where careful scans using microprobe analysis and energy and wavelength dispersive x-rays of cleaved surfaces proved the absence of metallurgical heterogeneities and impurities at the detection limit. High pressure studies showed the suppression of both ferromagnetism and signs of superconductivity at high pressure.

A more detailed study of the magnetization as a function of pressure recently revealed a surprising similarity of the ferromagnetic phase diagram of $ZrZn_2$ with that of UGe_2 [52, 53]. In these studies evidence for two metamagnetic transitions has been observed. Qualitatively the phase diagram of $ZrZn_2$ in the range from ambient pressure to 20 kbar appears to be identical with the phase diagram of UGe_2 in the pressure range 13 kbar $< p <$ 17 kbar. Since the characteristic width of f-bands is typically an order of magnitude smaller than that of d-bands, it is not surprising that pressures of the order $\sim$20 kbar have a much weaker effect in $ZrZn_2$. As for UGe_2, the phase diagram of $ZrZn_2$ may be explained by two peaks of the density of states that are related to van Hove singularities.

The general message in the context of superconductivity in the ferromagnets UGe_2 and $ZrZn_2$ is that novel forms of order may emerge at the border of first order quantum phase transitions for the same reasons as they were sought near quantum critical transitions. Yet the underlying dynamics may yield unexpected surprises, when a more quantitative analysis is carried out.

5. Summary

Discontinuous QPT are interesting for a number of reasons. First, discontinuous QPT may be very weakly first order, leaving behind an abundance of nearly quantum critical fluctuations that dominate the low lying excitations and drive the emergence of novel electronic states. In this limit first order quantum phase transitions are essentially like quantum critical points. Strict quantum criticality may then be stabilized with an external tuning parameter like magnetic field generating, for instance, a quantum critical end-point in materials exhibiting itinerant metamagnetism [53b]. Second, discontinuous QPT may also be the origin of novel types of low lying excitations. For instance, a first order free energy landscape may result in quantum instantons and the formation of metastable droplets that drive non-Fermi liquid phases [30]. Discontinuous QPT may finally signal the existence of subtle quantum correlation effects, such as magnetic rotons, the formation of skyrmion textures, non-analytic magnetization modes that result from a coupling to soft particle–hole excitations, or Pomeranchuk instabilities of the Fermi surface.

Remarkably, *all* quantum phase transitions in clean, stoichiometric compounds for which direct measurements of the order parameter exist to date, prove to be of first order. Examples include the emergence of superconductivity in band ferromagnets [41, 42] and at valence instabilities [54]. They also include superconductivity at the border of itinerant antiferromagnetism, for which a discontinuous suppression of antiferromagnetism has been suggested when $T_N < T_s$. Even non-Fermi liquid phases arise near first order behaviour. Empirically this may suggest that quantum critical points do not exist in real materials, except under very special circumstances. Yet, first order QPT promise to surprise by revealing further fascinating features in future studies.

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