Ferromagnetic quantum criticality

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During the last years strong efforts have been made in studying quantum criticality, in particular in ferromagnetic (FM) metallic systems. Substantial work was done in this Institute which has questioned theories developed in the late nineties and has helped to give a fresh boost to the field. The state of the art of this research area was summarized in a comprehensive review article. Most recently, we have found that Ce- and Yb-based Kondo-lattice ferromagnets order mainly along the magnetically hard direction of the ground-state Kramers doublet determined by crystalline electric field. This peculiar phenomenon, that was believed to be rare, is instead the standard case and can be explained by the so-called order-by-disorder theory. One exception to this is the strong anisotropic quasi-one-dimensional ferromagnet $CeRh_6Ge_4$. A systematic study of its properties under hydrostatic pressure has shown that a clean FM-QCP exists in this compound above which 'strange metallic behavior' was found. This was only previously seen in antiferromagnetic systems, and long thought to be impossible in a ferromagnet.

The general concept of quantum criticality has become one of the foundations for the study of strongly correlated electron physics. One of the main reasons for the broad interest of the scientific community in quantum critical points (QCPs), i.e., continuous (2^{nd}-order) phase transitions at zero temperature driven by quantum fluctuations instead of thermal fluctuations, is the presence of unconventional (non-phonon mediated) superconductivity found at and near these QCPs. This has been observed in various materials, like high-T_c superconductors, iron pnictides, organic metals or heavy-fermion systems despite their intrinsic difference in structure and physical properties. In fact, the common agreement is that strong quantum fluctuations present at these QCPs are the 'glue' for the electron pairing in the superconducting state [1].

The systems described above are mostly antiferromagnets. In ferromagnetic systems, however, superconductivity and QCPs are very rare. A long-standing question is whether a FM QCP can generally exist in metals and, if not, which are the possible ground states of matter that replace it. In recent years, substantial experimental and theoretical efforts have been made with groups from this Institute playing a major role. According to these recent studies it seems that a FM QCP can exist, but only under special circumstances. On theoretical grounds, it was shown that in 2D and 3D itinerant systems the quantum phase transition (QPT) from the paramagnetic (PM) to the ferromagnetic (FM) phase in the absence of quenched disorder is inherently unstable, either towards a first order phase transition or towards inhomogeneous magnetic phases (modulated or textured structures). This conclusion, proposed in the late 1990s [2] has been confirmed by different theoretical approaches and several experimental studies [3]. The generic phase diagram of these systems is shown in Fig. 1a in the space spanned by temperature *T*, magnetic field *H* and the control parameter pressure *p* or chemical substitution *x*. In quasi-1D systems, however, FM QCPs can exist, as has been demonstrated in our study on $YbNi_4(P_{1-x}As_x)_2$ in which As substitution acts as negative pressure [4]. The phase diagram of this system is shown in Fig. 1b. On the other hand, disordered systems are much more complicated, depending on the disorder strength and the distance from the QCP. In many disordered materials the phase diagram shows a 'tail' near the putative QCP, as shown

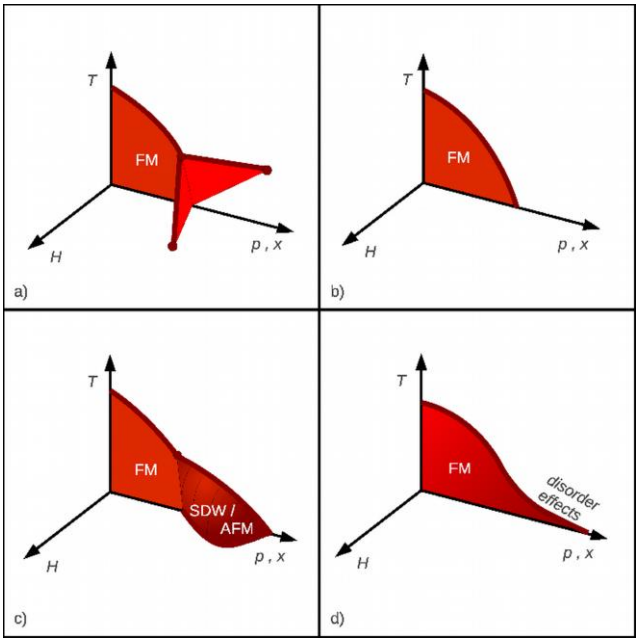

Fig. 1: Generic phase diagrams observed in metallic ferromagnets in the space spanned by temperature, T, magnetic field, H, and the control parameter pressure p or chemical substitution x. Figure taken from Ref. [4].

in Fig. 1d, and a transition in to a state with glass-like spin dynamics is suspected [5].

In other systems the transition from the FM state at low temperatures is to a different type of long-range order, such as an antiferromagnetic (AFM) or a spin-density-wave state (SDW). The phase diagram of these systems can schematically be represented as in Fig. 1c. Examples discovered and studied by groups in our Institute include *3d*-electron systems, like $NbFe_2$ [5] or *4f*-electron systems like CeRuPO [6] or $CeTi_{1-x}V_xGe_3$ [7].

Recently, we have presented a detailed study of the evolution of the magnetic order in $Yb(Rh_{1-x}Co_x)_2Si_2$ from a FM state with moments along the very hard *c* direction at $x = 0.27$ towards the yet unknown magnetic state at $x = 0$ [8]. We first observe a transition towards an AFM canted state with decreasing *x* and then to a pure AFM state. This confirms that the QCP in $YbRh_2Si_2$ is AFM, but the phase diagram is very similar to those observed in the inherently FM systems like $NbFe_2$ and CeRuPO, which suggests that the basic underlying instability might be FM. Despite the huge CEF anisotropy the ordered moment retains a component along the *c* axis also in the AFM state. The phase diagram in zero field is shown in Fig. 2.

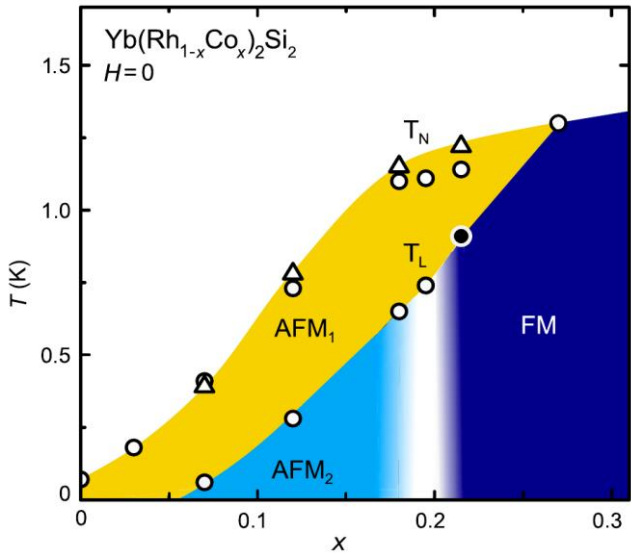

Fig. 2: Phase diagram of $Yb(Rh_{1-x}Co_x)_2Si_2$. The ferromagnetic (FM) phase is separated from two antiferromagnetic phases, AFM_1 and AFM_2, by first order lines. The filled point indicates a first order transition. Since we have investigated only samples with $x = 0.18$ and 0.21, we do not know the exact location of the line between the FM and the AFM_2 phases and we left this area uncolored. Figure taken from Ref. [8].

Our results have a further important consequence: The phase boundary line between the AFM_2 phase and the PM phase is first order and terminates at a multicritical point (MCP) at finite temperature. If this point was shifted to $T = 0$ at a certain concentration between 0.12 and 0, then it would have the nature of a field-induced quantum MCP in remarkable agreement with predictions of Misawa et al. [9] and very similar to what has been observed in $NbFe_2$ [5].

Another intriguing property of this system is the occurrence of magnetic order along a very hard crystalline electric field (CEF) direction, i.e., along the one with the smallest available magnetic moment. The huge CEF anisotropy in $Yb(Rh_{1-x}Co_x)_2Si_2$ excludes that this hard-axis ordering originates from a competing exchange anisotropy as often proposed for other heavy-fermion systems. Instead, it points to an order-by-disorder based mechanism [10].

This property, however, has not only been observed in this system but in several other FM Kondo lattices (KLs) [11], so that we have performed a detailed study of this effect [12]. This is exemplarily shown in Fig. 3 in which we have plotted the temperature dependence of the ac-susceptibility $\chi'(T)$ of four KL systems, $YbNi_4P_2$, $Yb(Rh_{0.73}Co_{0.27})_2Si_2$, CeRuPO, and $YbIr_3Ge_7$, measured with magnetic field along the two principal crystallographic directions. At high temperatures, both susceptibilities follow the same *T* dependence, because of the dominant Curie-Weiss contribution of the full moment of trivalent Ce and Yb. However, their absolute values differ significantly due to the magnetocrystalline anisotropy caused by the CEF of the tetragonal structure. At low temperatures, just above T_C, these susceptibilities cross each other at a temperature T_0 (marked by an arrow in the first panel of Fig. 3), which inevitably indicates that the magnetic moments order along the magnetically hard direction of the CEF. Below T_C, the measured $\chi'(T)$ perpendicular to the ordered moments remains constant, while the behavior of $\chi'(T)$ parallel to the moments depends on the ratio between the coercive field and the modulated field used in the measurements: For instance, in YbNi4P2, $\chi'(T)$ stays constant below T_C, whereas in $Yb(Rh_{0.73}Co_{0.27})_2Si_2$ it decreases steeply. The fact that T_0 is just above T_C implies that there is no correlation between this behavior and the CEF first excited state which is located at much higher temperatures in these systems.

TABLE I. Ce- and Yb-based Kondo-lattice ferromagnets. YRCS=$Yb(Rh_{0.73}Co_{0.27})_2Si_2$, T_C=Curie temperature, T_K=Kondo temperature, μ=magnetic moment, CEF=crystalline electric field, ESR=electron spin resonance (9.4 GHz, $T > 3$ K, main crystalline directions), MFT=mean-field transition, GS=ground-state wave function, NA=not available, *=moments order along the CEF magnetically hard axis.

System	T_C (K)	T_K (K)[a]	Cryst. structure	Ordered μ (μ_B)	CEF anisotropy	Easy axis	Order axis	ESR signal	MFT	Coherence max. in $\rho(T)$
*$CeAgSb_2$ [34–39]	9.6	16	Tetra.	0.41	3	*ab* plane	*c* axis	Not found	Weak	Yes
*CeRuPO [21,40,41]	15	7	Tetra.	0.3	3	*ab* plane	*c* axis	Found	No	Yes
*$CeFeAs_{0.7}P_{0.3}O$ [42]	7.5	5	Tetra.	0.3	3	*ab* plane	*c* axis	Found	No	Yes
*YRCS [17,33,43]	1.3	7.5	Tetra.	0.1	6	*ab* plane	*c* axis	Found	No	Yes
*$YbNi_4P_2$ [12–14,44][b]	0.15	8	Tetra.	<0.05	5	*c* axis	*ab* plane	Not found	No	Yes
*$YbIr_3Ge_7$ [22]	2.6	16	Rhomb.	0.05	4	*c* axis	*ab* plane	Not found	No	Yes
*YbNiSn [16,45,46]	5.6	10	Orth.	0.85	1.8	*a* axis	*c* axis	Not found	No	Yes
*YbPtGe [47]	5.4	9.4	Orth.	1	2	*a* axis	*c* axis	NA	No	Yes
*YbRhSb [48,49][c]	4.3	30	Orth.	0.4	2	*a* axis	*c* axis	NA	No	Yes
*YbPdSi [30][d]	8	13	Orth.	0.26	NA	*b* axis	*c* axis	NA	No	Yes
*β-$CeNiSb_3$ [31][e]	6	10	Orth.	0.9	1.5	*b* axis	*c* axis	NA	No	Yes
*$CeIrGe_3$ [50–52][f]	4.8	12	Tetra.	0.14	1.3	*a* axis	*c* axis	NA	First order	Yes
$CeTiGe_3$ [25]	14	30	Hexag.	1.5	10	*c* axis	*c* axis	Not found	Yes	Yes
$CeRu_2Al_2B$ [26,27,53][g]	13	23	Tetra.	1	>40	*c* axis	*c* axis	NA	NA	Yes

[a]Reported or estimated from entropy.
[b]In-plane anisotropy: orthorhombic point symmetry site for Yb.
[c]Under pressure of about 2 GPa. At zero pressure the order is canted AFM with a very small ordered moment ($0.003\mu_B$ along the *b* axis). T_K is the value at zero pressure.
[d]Largest moment in a complex structure with three Yb sites and three different moment sizes.
[e]Two Ce sites.
[f]Transition into a canted AFM at 8.7 K, which is probably first order. Recent neutron experiments suggest the magnetic structure is more complex than a collinear FM [52].
[g]AFM transition at 14.3 K.

Eventually, we found that almost all Ce- and Yb-based Kondo-lattice ferromagnets order mainly along the magnetically CEF hard direction. A list of those materials is given in Table I copied from Ref. [12]. This behavior is independent of the Curie temperature T_C, crystalline structure, size of the ordered moment, and type of ground state wave function. On the other hand, all these systems show Kondo temperatures of a few degrees Kelvin, often close to T_C, and they have in common a relatively small CEF anisotropy. CEF excited states are too high in energy to be responsible for this behavior. Specific heat measurements indicate that the second-order phase transition is not mean-field-like, pointing to an important role of fluctuations, which might induce such an order along the hard axis. However, the intrinsic mechanism leading to this kind of order in all KL ferromagnets remains unknown. We further note that a huge Ising-type anisotropy prevents this unexpected type of ordering and leads to conventional order along the easy axis.

This is, in fact, the case of a recently discovered strong anisotropic quasi-one-dimensional ferromagnet $CeRh_6Ge_4$ (T_C = 2.5 K). Together with our collaborators, we found that the pristine heavy fermion ferromagnet $CeRh_6Ge_4$ exhibits a ferromagnetic instability under the 'clean' tuning parameter, hydrostatic pressure [13]. A few years ago, we have observed evidence of a ferromagnetic instability in the substituted heavy fermion metal $YbNi_4(P_{1-x}As_x)_2$ [4].

However, in this study elemental substitution served as tuning parameter and the ferromagnetic instability could be simulated by the alloying-induced disorder.

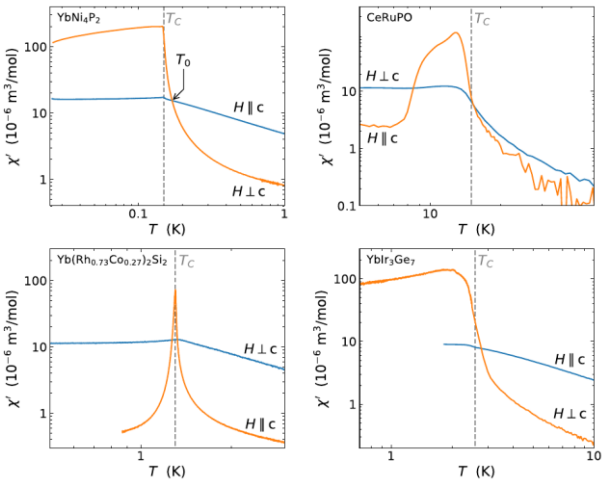

Fig. 3: Temperature dependence of the magnetic ac-susceptibility $\chi(T)$ for $YbNi_4P_2$, $Yb(Rh_{0.73}Co_{0.27})_2Si_2$, CeRuPO, and $YbIr_3Ge_7$ measured with modulated field along the two principal CEF directions (H_{ac} // c and H_{ac} ⊥ c) of the tetragonal crystalline structure. Strong noise is seen for CeRuPO due to the very small size of the crystal. Figure taken from Ref. [9].

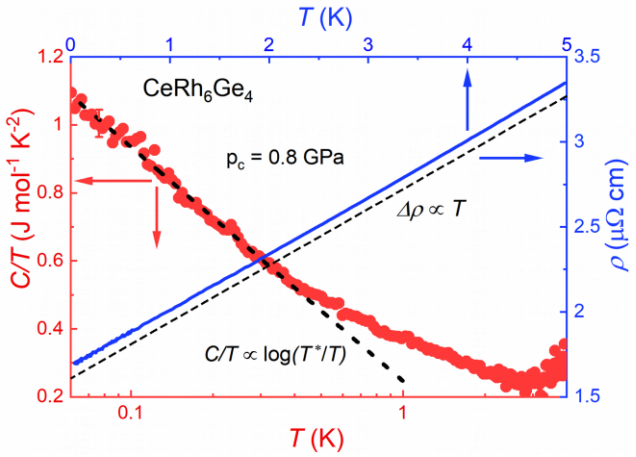

Fig. 4: Electrical resistivity ρ(T) and specific heat C(T)/T at p_c = 0.8 GPa. ρ(T) exhibits linear behavior extending from 5 K down to at least 40 mK, whereas C(T)/T shows continues to increase logarithmically with decreasing temperature. Figure taken from Ref. [13].

On the contrary, that is not the case upon pressurizing $CeRh_6Ge_4$.

The application of hydrostatic pressure to single crystals of $CeRh_6Ge_4$ suppresses the ferromagnetic order to absolute zero as shown in the phase diagram in Fig. 5. Near this quantum critical point at p_c = 0.8 GPa, $CeRh_6Ge_4$ develops the 'strange metallic behavior' only previously seen in AFM systems, and long thought to be impossible in a ferromagnet. The strange metallic behavior is evidenced by a logarithmic

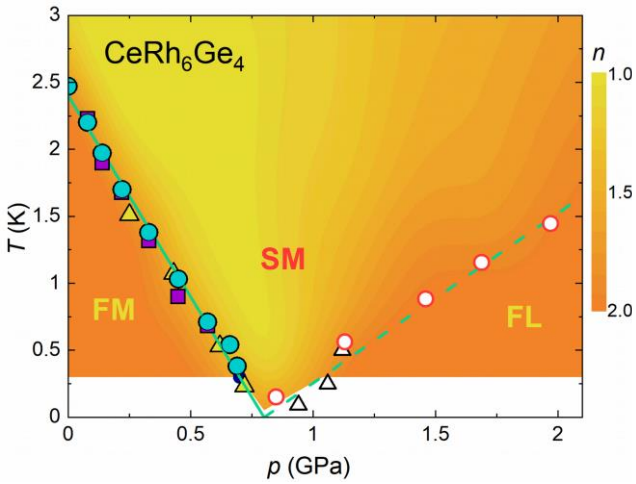

Fig. 5: T−p phase diagram of $CeRh_6Ge_4$. The circles, triangles and squares for pressures below $p_c \approx 0.8$GPa denote T_C derived from the resistivity and specific heat. The corresponding symbols above p_c mark TFL, below which Fermi-liquid behavior occurs. The ferromagnetic (FM), the strange metal (SM), and the Fermi-liquid (FL) phases are indicated. The color map corresponds to the resistivity exponent n. Figure taken from Ref. [13].

diversion of the specific heat and a linear temperature dependence in the electrical resistivity displayed in Fig. 2. An important clue to this behavior is that $CeRh_6Ge_4$ is extremely magnetically anisotropic, a property which not only restricts the direction of the ferromagnetism to a plane but in doing so, also injects entangled electron spin pairs into the ferromagnet. This result confirms the role of electron entanglement as a key element of the strange metal. The fact that this system has a non-centrosymmetric crystalline structure seems to play a fundamental role in explaining the presence of the FM-QCP [14].

External Cooperation Partners

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