Advancing quantum criticality

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During the last years strong efforts have been made in studying quantum criticality, in particular in ferromagnetic 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 has now been summarized in a comprehensive review article. Most recently, the focus of our research was shifted to other types of quantum criticality which include quantum multi-criticality, peculiar charge-density-wave quantum criticality, which promotes superconductivity and also complex multipolar quantum criticality, a poorly explored field in which competition between orbital and spin degrees of freedom control the ground state of the system at zero temperature.

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 unconven-tional (non-phonon mediated) superconductivity found at and near these QCPs. This has been observed in various materials, like high-T_c super-conductors, 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].

Ferromagnetic quantum criticality

The systems described above are mostly antiferro-magnets. In ferromagnetic (FM) 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 para-meter 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 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

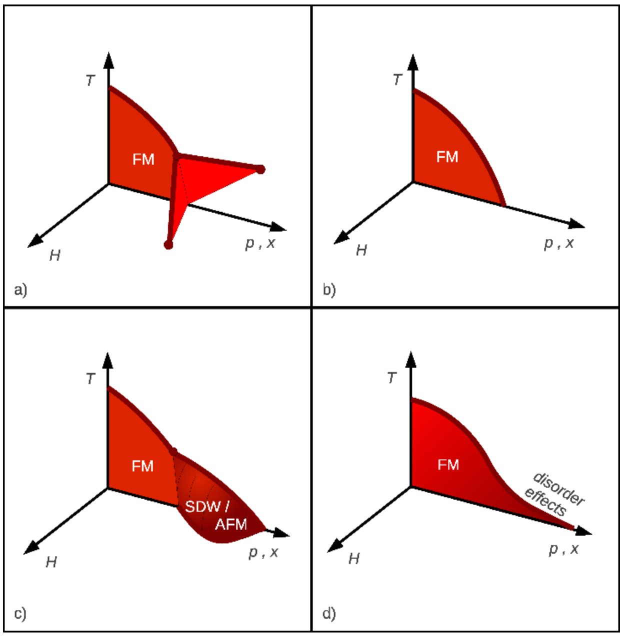

Fig.-1: Generic phase diagrams observed in metallic ferromagnets in the space spanned by temperature, T, magnetic field, H, and the control parameter pressure p or chemical substitution x (Figure taken from Ref. 3).

can schematically be represented as in Fig. 1c. Examples discovered and studied by groups in our Institute include *3d*-electron systems, like $NbFe_2$ [6] or $TaFe_2$ [7], as well as *4f*-electron systems like CeFePO or CeRuPO. Recently, we have shown that chemical substitution with As or Ru drives the latter systems from a FM to an AFM state [8]: The phase diagram in zero field is shown in Fig. 2.

Quantum multi-criticality

Ferromagnetic quantum critical systems of the last kind are maybe the most exciting, because in some points of their phase diagram the presence and even divergence of both AFM and FM quantum fluctuations is allowed. These points are quantum tri-critical points (QTCPs) in the simplest case, i.e., points in which three phase lines terminate, but could be multi-critical if more phase lines converge. A similar situation has been recently discussed for the layered perovskite metal $Sr_3Ru_2O_7$ which is a paramagnetic metal close to a FM QCP and presents an unusual phase diagram in which two SDW phases are induced by a magnetic field through a QCP and two 1^s-order phase transitions [8].

The existence of such QTCPs has been shown in one of these materials, the relatively simple binary metallic magnet $NbFe_2$ [10]. In this compound, a change from a low-temperature FM state in an incommensurate SDW state under small chemical pressure was found [6], blue and red areas in Fig. 3, respectively. If a magnetic field is applied along a particular crystallographic direction while the system is in the SDW state, a 1^{st}-order phase transition into the PM state can be induced, indicated by grey lines in Fig. 3. These lines end at tri-critical points (TCP) (small red ball in Fig. 3). We have followed and investigated the properties of the TCP to zero temperature, the quantum tri-critical point, marked by the orange balls in Fig. 3. We successfully used a well-known two-order-parameter Landau theory to understand the phase diagram of $NbFe_2$, which is considered to be generic for all systems with an avoided FM QCP due to an adjacent AFM phase like in CeRuPO or, as we recently found, in the prominent heavy-fermion compound $YbRh_2Si_2$.

Charge-density-wave quantum criticality

While QCPs found in magnetic materials are quite common and has been intensively investigated, QCPs at structural phase transitions are very rare. This is mainly due to the fact that structural phase transitions are typically 1^{st} order. However, in a few particular cases, like in the quasi-skutterudite superconductor $Sr_3Rh_4Sn_{13}$, a high-temperature 2^{nd}-order structural phase transition can be tuned to $T = 0$ by hydrostatic pressure or chemical substitution with Ca for Sr. Here, superconductivity appears at low temperature and persists across the whole pressure range increasing only weakly around the QCP [11].

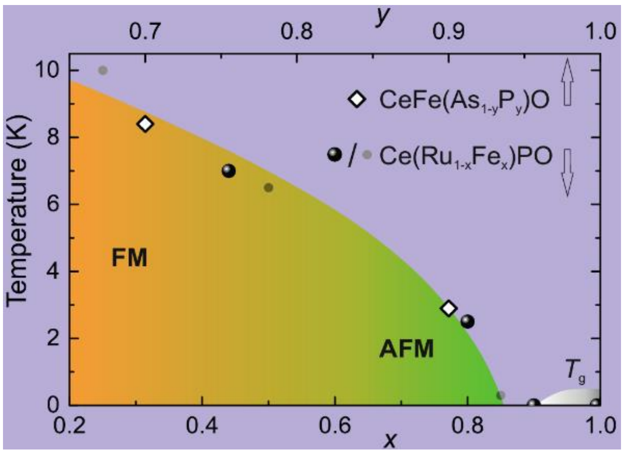

Fig.-2: Temperature–substitution phase diagram of the series $Ce(Ru_{1-x}Fe_x)PO$ and $CeFe(As_{1-y}P_y)O$ (Figure taken from Ref. 8).

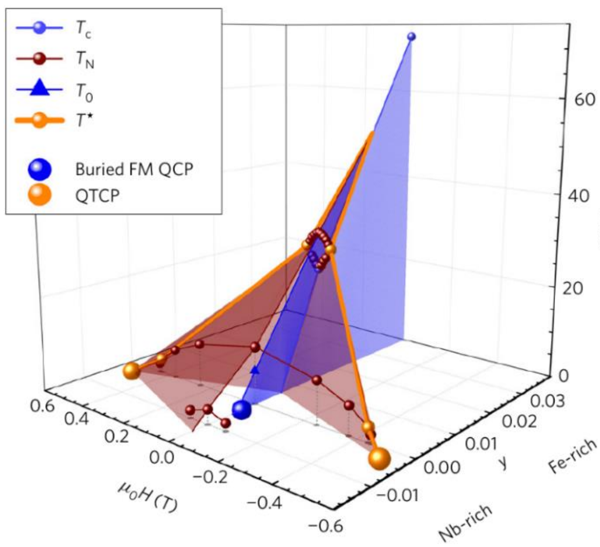

Fig.-3: Overall composition-magnetic field-temperature phase diagram for the $Nb_{1-y}Fe_{2+y}$ system. The underlying ferromagnetic transition temperature T_0 is extracted from the data analysis of the magnetization M which assumes a Landau expansion of the free energy up to the M^4 term. The phase boundaries of the SDW (red) and FM (blue) phase are obtained from magnetization and sus-ceptibility measurements. The position of the avoided ferromagnetic QCP (blue ball) and the QTCPs (orange ball) are highlighted (Figure taken from Ref. 10).

During studies of compounds with cubic Heusler $L2_1$-type structures [12] we discovered that $LuPt_2In$ displays a 2nd-order structural transition of charge-density-wave (CWD) type at T_{CDW} = 490 K and that is superconducting below T_c = 0.4 K. It was possible to tune this temperature to zero by substitution of Pd for Pt. The QCP was found to be at the critical palladium content x_c = 0.58 [13]. This is illustrated in the upper panel of Fig. 4. Surprisingly, we observed a strong increase in T_c which at the QCP is almost double of the value at $x = 0$, and the disappear-ance of superconductivity at higher Pd content. Although the electron pairing is phonon-mediated (no magnetic elements are present in this system), the presence of a pronounced peak in T_c at exactly the CDW QCP is a unique feature among quantum critical CDW systems and points to a new type of interaction between the CDW and superconductivity. This study and recent developments indicate that superconductivity might be mediated by a soft mode associated to quantum criticality. We believe that this discovery is of major importance, since it is related to other relevant systems like the high-Tc superconductors, in which, on the other hand, the CDW seems to compete with superconductivity.

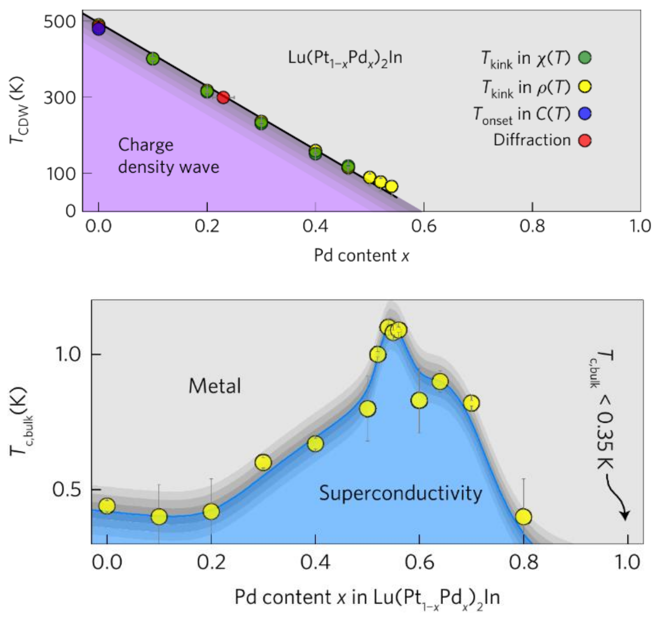

Fig.-4: Upper panel: The temperature-composition phase diagram of $Lu(Pt_{1-x}Pd_x)_2In$. T_{CDW} values were determined in different experiments and show a linear decrease with increasing Pd content. The linear extrapolation of T_{CDW} to $T = 0$ indicates a CDW QCP at $x_c = 0.58$. Lower panel: Composition dependence of T_c which presents a sharp peak at the QCP: the fast decrease of T_c for $x > x_c$ is very different from the weak dependence usually observed in CDW systems (Figure taken from Ref. 13).

Multipolar quantum criticality

Another area of research that is becoming more significant is the study of multipolar systems and their quantum criticality. One of the motivating factors is the recent discovery of heavy-fermion superconductivity across the ferro-quadrupolar QCP of $PrTi_2Al_{20}$ [14]. Another factor is that not much is known about the behavior of multipolar systems near QCPs. This is mainly due to the scarcity of materials which show multipolar order and the experimental difficulties to magnetic measurements. On the other hand, the interplay between magnetic and orbital degrees of freedom is appealing, although this makes the phase diagram of such materials very complex.

A prototypical example of such a complex system is the cubic metal CeB_6. Although simple in its struc-ture, it shows at zero field antiferro-quadrupolar (AFQ) order below T_Q = 3.3 K and at even lower temperatures AFM order below T_N = 2.4 K. If cerium is substituted by lanthanum a new phase appears, which is believed to be an octupolar (AFO) phase. All this variety of states of matter becomes more complex if the system is tuned by a magnetic field which can induce a magnetic dipole moment.
In our recent study of the phase diagram through thermodynamic measurements, we have revisited and corrected the phase diagram of $Ce_{1-x}La_xB_6$, which is shown in Fig. 5 [15]. More importantly, we have identified the presence of a QCP associated to multipolar quantum critical fluctuations near $x = 0.75$ which is directly correlated to the effective electron mass. This is shown by the high values of the Sommerfeld coefficient at the multipolar QCP (see red area in Fig. 5).

Another exciting new superconductor which has been recently discovered in our Institute is the tetragonal $CeRh_2As_2$. This system has a peculiar quasi-quartet ground state which allows quadrupolar order. If confirmed, it would be the first case of a Ce-based system near a quadrupolar QCP which shows heavy-fermion superconductivity.

External cooperation partners

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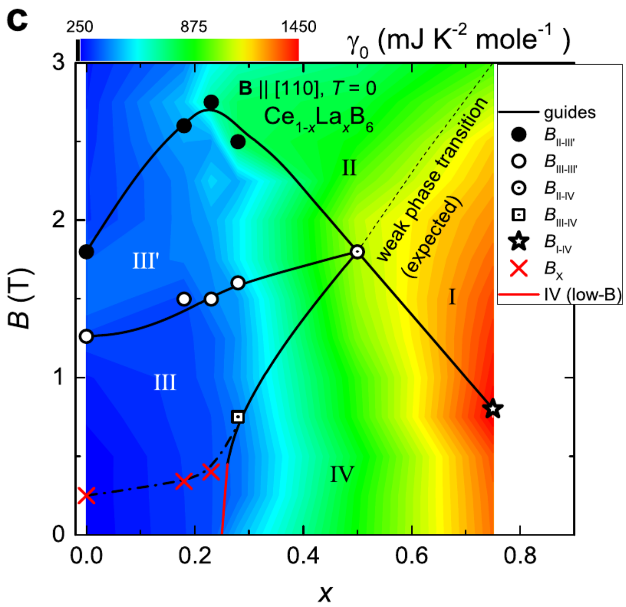

Fig.-5: Composition (x) - magnetic field (B) phase diagram of $Ce_{1-x}La_xB_6$ at T = 0. By convention the phases for the primary order parameters, the spin/pseudospin (orbital) paramagnetic (PM) phase, the antiferro-quadrupolar (AFQ) phase, the antiferromagnetic (AFM) phase, and the antiferro-octupolar (AFO) phase are referred to as phases I, II, III, and IV, respectively. The red segment for the low-B III–IV phase boundary is to emphasize the large increase of the Sommerfeld coefficient γ_0 upon entering the phase IV. The background is a contour plot of $\gamma_0(x,B)$ which has its maxima values near the QCP at $B_{I\text{-}IV}$ (Figure taken from Ref. 15).

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References

[1] *Superconductivity without phonons, P. Monthoux, D. Pines and G. G. Lonzarich, Nature **450** (2007) 1177.*

[2] *First Order Transitions and Multicritical Points in Weak Itinerant Ferromagnets, D. Belitz, T. R. Kirkpatrick and T. Vojta, Phys. Rev. Lett. **82** (1999) 4707.*

[3]* *Metallic quantum ferromagnets, M. Brando, D. Belitz, F. M. Grosche and T. R. Kirkpatrick, Rev. Mod. Phys. **88** (2016) 025006.*

[4]* *Ferromagnetic Quantum Critical Point in the Heavy-Fermion Metal $YbNi_4(P_{1-x}As_x)_2$, A. Steppke, R. Küchler, S. Lausberg, E. Lengyel, L. Steinke, R. Borth, T. Lühmann, C. Krellner, M. Nicklas, C. Geibel, F. Steglich and M. Brando, Science **339** (2013) 933.*

[5] *Towards ferromagnetic quantum criticality in $FeGa_{3-x}Ge_x$: ^{71}Ga NQR as a zero-field microscopic probe, M. Majumder, M. Wagner-Reetz, R. Cardoso-Gil, P. Gille, F. Steglich, Y. Grin and M. Baenitz, Phys. Rev. B **93** (2016) 064410.*

[6]* *Spectroscopic study of metallic magnetism in single-crystalline $Nb_{1-y}Fe_{2+y}$, D. Rauch, M. Kraken, F. J. Litterst, S. Suellow, H. Luetkens, M. Brando, T. Foerster, J. Sichelschmidt, A. Neubauer, C. Pfleiderer, W. J. Duncan and F. M. Grosche, Phys. Rev. B **91** (2015) 174404.*

[7]* *Quantum Phase Transitions and Multicriticality in $Ta(Fe_{1-x}V_x)_2$, M. Brando, A. Kerkau, A. Todorova, Y. Yamada, P. Khuntia, T. Förster, U. Burkhard, M. Baenitz and G. Kreiner, J. Phys. Soc. Jpn. **85** (2016) 084707.*

[8]* *Avoided ferromagnetic quantum critical point: Antiferromagnetic ground state in substituted CeFePO, A. Jesche, T. Ballé, K. Kliemt, C. Geibel, M. Brando and C. Krellner, Phys. Status Solidi **1** (2017) 1600169.*

[9]* *Low temperature thermodynamic investigation of the phase diagram of $Sr_3Ru_2O_7$, D. Sun, A. W. Rost, R. S. Perry, A. P. Mackenzie and M. Brando, Phys. Rev. B **97** (2018) 115101.*

[10]* *Quantum tricritical points in $NbFe_2$, S. Friedemann, W. J. Duncan, M. Hirschberger, T. W. Bauer, R. Küchler, A. Neubauer, M. Brando, C. Pfleiderer and F. M. Grosche, Nat. Phys. **14** (2018) 62.*

[11] *Ambient Pressure Structural Quantum Critical Point in the Phase Diagram of $(Ca_xSr_{1-x})_3Rh_4Sn_{13}$, S. K. Goh, D. A. Tompsett, P. J. Saines, H. C. Chang, T. Matsumoto, M. Imai, K. Yoshimura and F. M. Grosche, Phys. Rev. Lett. **114** (2015) 097002.*

[12]* *Unusual weak magnetic exchange in two different structure types: $YbPt_2Sn$ and $YbPt_2In$, T. Gruner, D.-J. Jang, A. Steppke, M. Brando, T. Ritter, C. Krellner and C. Geibel, J. Phys.: Condens. Matter **26** (2014) 485002.*

[13]* *Charge density wave quantum critical point with strong enhancement of superconductivity, T. Gruner, D.-J. Jang, Z. Huesges, R. Cardoso-Gil, G. H. Fecher, M. M. Koza, O. Stockert, A. P. Mackenzie, M. Brando and C. Geibel, Nat. Phys. **13** (2017) 967.*

[14] *Pressure-Induced Heavy Fermion Superconductivity in the Nonmagnetic Quadrupolar System $PrTi_2Al_{20}$, K. Matsubayashi, T. Tanaka, A. Sakai, S. Nakatsuji, Y. Kubo and Y. Uwatoko, Phys. Rev. Lett. **109** (2012) 187004.*

[15]* *Large positive correlation between the effective electron mass and the multipolar fluctuation in the heavy-fermion metal $Ce_{1-x}La_xB_6$, D.-J. Jang, P. Y. Portnichenko, A. S. Cameron, G. Friemel, A. V. Dukhnenko, N. Y. Shitsevalova, V. B. Filipov, A. Schneidewind, A. Ivanov, D. S. Inosov and M. Brando, npj Quantum Materials **2** (2017) 62.*

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