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Present Status and Newroute in Heavy Fermion Physics

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A brief summary is given on the development of heavy fermion physics. The main aim is to push to new instrumental improvements. Focus is made on the possibility to realise a new generation of clean experiments in extreme conditions (P, T, H). Three examples illustrate the future: $CeRu_2Ge_2$ with the feedback between magnetic interaction, Fermi surface and the localisation of the 4f electron, $CePd_2Si_2$ with the collapse of antiferromagnetism and emergence of superconductivity and SmS with the goal in its gold phase to follow the pressure collapse of its many body insulating ground state and the appearance of long range magnetic ordering.

KEYWORDS: heavy fermion, quantum critical point (QCP)

§1. Evolution of the Subject: from Intermediate Valence to Magnetic Quantum Critical Point

The physics of heavy fermions may start with the temperature - pressure, phase diagram (T, P) of pure metallic cerium with the so called γ - α first order transition line between the high temperature γ phase and the low temperature α phase.[1)] At the $T_{\gamma\alpha}$ line the transition is characterised by an isostructural collapse of the volume ($\sim 10\%$) associated with a slight increase of the cerium valence. At $P = 0$ (Fig. 1), $T_{\gamma\alpha}(0) \sim 100$ K is rather large so no discussion has been made on the possible occurence of long range magnetism.[2,3)] In the α phase, the weak intermediate valent character of the Ce ions corresponds to a large Kondo temperature (T_K) which prevents any long range magnetic ordering.

The discovery of the heavy fermion compound $CeAl_3$[4)] by the detection of huge values of the extrapolated γ=1.6 J.mole^{-1}K^{-2} coefficient of the linear temperature contribution of the specific heat $C = \gamma T$ at $T = 0$ K and of the A coefficient of the T^2 term of the resistivity really raises the question of the competition between the paramagnetic state (PA) and long range magnetic ordering, ie the location of the magnetic quantum critical point (QCP) where the ordering temperature T_N or T_C for antiferromagnetic (AF) or ferromagnetic (F) phase collapses. For the well ordered antiferromagnetic compound $CeAl_2$ ($T_N \sim 3.8$ K at $P = 0$), a γ value as large as 120 mJ.mole^{-1}K^2 ie two order of magnitude bigger than that of metallic copper has been measured below T_N.[5,6)] The heavy fermion character occurs for (mostly) AF or F systems near a QCP. It is worthwhile to remark that if measurements are only performed above 10 K, it cannot be predicted which compounds in the Ce, Al family ($CeAl_2$, Ce_3Al_{11}, $CeAl_3$) will choose between AF or PA states.[7)] The disappearance with pressure of T_N in $CeAl_2$ near 30 kbar[8)] seems to agree well with the Doniach picture[9)] that, under pressure, T_N collapses as the local Kondo fluctuation overcomes the intersite Rudermann Kittel Kasuya Yoshida interaction between the localised magnetic moments.

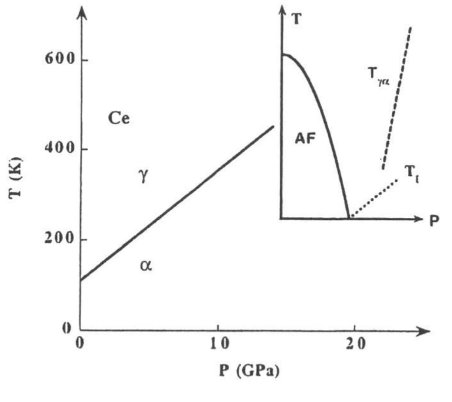

Fig. 1. T, P phase diagram of metallic cerium. The continuous line is the first order $T_{\gamma\alpha}(P)$ transition line: long range magnetic ordering cannot appear in the intermediate valence α phase. The insert shows the opposite situation where antiferromagnetic ground state occurs below the QCP at $P = P_c$; Fermi liquid properties in the paramagnetic state can be observed only below T_I.

Near the QCP, as the magnetism is very sensitive to pressure, uniaxial stress or defects, the intrinsic properties can be quite tricky and difficult to identify. For more than a decade, $CeAl_3$ was the archetype example of a paramagnetic heavy fermion ground state; maxima in temperature of C/T and negative minima of the thermal expansion were taken as coherent temperature T^*.[7)] The growth of single crystals allows a new generation of experiments which show clearly long range magnetic order.[10,11)] The weak temperature maximum of C/T measured previously on polycrystals appears now as a consequence of a broadened distribution of T_N. A maximum of $\gamma = (C/T)_{T\to 0}$ occurs for $P_c \sim 2$ GPa;[12,13)] here the temperature variation of C/T shows a continuous decrease (no maximum) on warming as predicted by any spin fluctuation theory for nearly AF or F systems ($P \geq P_c$).[14]

For many years, the study of the low temperature paramagnetic domain was mainly limited to cases rather far from the QCP, the temperature dependence of observables like specific heat was poorly analysed. The fashion was mainly to scale by a unique parameter ie a characteristic temperature reminiscent of the Kondo scaling. The focus was concentrated on unconventional superconductors after their discovery in $CeCu_2Si_2$,[15] UBe_{16}[17] and UPt_3;[18] special attention was paid on the double superconducting transition of UPt_3.[14] Another important class of investigation concerns the many body insulator as observed for example in the low pressure phase of the intermediate valent compound like SmS as discussed below.[19] Of course, the success of quantum oscillation techniques in heavy fermion compounds was decisive[20,21] to determine the Fermi surface. They confirm the validity of band calculation, and demonstrate the local or itinerant character of the f electron and the importance of electronic correlations in the heavy mass formation. The full Fermi surface with an f itinerant character has been determined in the low field phase of UPt_3 and $CeRu_2Si_2$ while in the ferromagnetic case of $CeRu_2Ge_2$ by contrast the f electron seems localised.[22,23]

§2. Magnetic Quantum Critical Point: Just above in the Paramagnetic Regime

The interest to revisit the magnetic QCP may first come from theoretical previsions that, for two channel Kondo effect, non Fermi liquid properties can be expected at low temperature.[24] In fact, a large crossover in temperature often referred as non Fermi liquid behaviour is observed in Ce lattices near a QCP; no two channel Kondo effects can be involved as the crystal field ground state is often a doublet. Theoretically, the problem is not new as it is well known that, for itinerant electrons near a QCP, very low energy fluctuations are involved. As it is well explained in the lecture notes of Ph. Nozières[25] for a ferromagnetic instability, all the crossover temperatures (ie temperature T_I below which Fermi liquid properties appear and temperature T_{II} above which high temperature law is recovered) are only a consequence of the sharp frequency and wavelength dependence of the dynamical susceptibility. Just at the QCP, the Fermi liquid will collapse, the ratio T_I/T_{II} diverges and so a large crossover regime is expected. Qualitatively, that is the experimental observations however the striking point is that, contrary to the prediction of the spin fluctuation theory in an ideal lattice,[14,26] well defined temperature laws such as a $LogT$ dependence of C/T are observed over more than a temperature decade [27]. There is as yet no explanation of a large crossover where a unique temperature dependence is observed. The interplay of disorder and spin fluctuations in the resistivity (ρ) seems to explain the T^α law ($\alpha \sim 1$) observed near a QCP; the physical reason is the enhancement of the scattering on Fermi surface along hot lines connected by the AF wavevector $\mathbf{Q}$.[28]

The interest of heavy fermion compounds is to reach the QCP at a rather moderate pressure (P_c = 20 kbar) to explore a large range of temperature ($T \leq T_I$ and $T \geq T_{II}$) to also allow studies of a highly electronic magnetic polarised phase with rather moderate field ($H_M \sim 10$ T). Furthermore the material is often not fragile, large crystals can be grown and excellent electrical contacts via spot welding can be achieved. Due to the low energy scale, the heavy fermion contribution is large in observables like ρ or C; that allows quantitative studies under external variables like P or uniaxial stress. Depending of the relative strength of the single Kondo temperature T_K, the exchange coupling J_Q and their anisotropy, the heavy fermion systems are quite varied. In the $CeCu_6$ family, the exchange J_Q has a wide wavevector distribution, while in the $CeRu_2Si_2$ serie J_Q is far more peaked in wavevector.[29] For this latter system, AF correlations observed by neutron scattering appear already below 70 K.[30] To speak about a single Kondo temperature of 20 K is clearly inappropriate.[31]

At zero pressure, the tuning through a QCP is achieved often by doping. This always leads to the difficulty to separate intrinsic effects and extrinsic phenomena correlated with the concentration distribution always present in alloys. The duality between the localised and itinerant character of the f electrons may lead to high sensitivity to defects as dislocation or stacking faults which correspond to huge pressure gradients. That may nucleate clusters with a magnetic collapse (local QCP) shifted by comparisons to the ideal lattice case. This non uniformity can be the cause of the so-called Griffith phase description which is now very popular.[32]

§3. Just below the QCP

At present, focus has been given generally to the paramagnetic behaviour just above the QCP; looking at all published data it is surprising that only crude analysis have been made in the AF domain. For the two substitutes series $CeCu_{6-x}Au_x$,[27] $Ce_{1-x}La_xRu_2Si_2$[33] below the QCP the common striking point is the rounding of the specific heat anomaly by contrast to the prevision of a molecular field behavior as given by the spin fluctuation approach. For example, the specific heat anomaly at T_N must correspond to a jump $\frac{\Delta C}{T_N} \sim T_N$, while the gaussian fluctuations disappear rapidly as $T_N \rightarrow 0$.[34] A deeper study of the magnetic boundary is crucial. In fact, recent NMR experiments realised on MnSi are quite illustrative of discoveries found by microscopic experiments. During a long period, MnSi seems to be one of the best example of ferromagnetic systems; it seems to end up in a PA ground state above $P_c = 15$ kbar through a second order phase transition.[35] The first indication of a more complex situation appears with the detection of tiny irreversibility in the susceptibility at the curie temperature, T_C, for $P \geq 12$kbar[36] and the second with the observation of a field reentrant long range magnetism.[37] NMR experiments[38] show a clear first order transition between the magnetic and paramagnetic phase as there is a finite discontinuity of the Si NMR frequency to zero at P_c.

§4. New Route: Continuous Sweep of Hydrostatic Pressure

To test further the theory, the disorder must be reduced as much as possible. The elegant way is to realise pressure experiments in a controlled way ie high hydrostaticity and continuous tuning.[39] The goal is to approach, in electronic systems, the same quality of experiments as those achieved on quantum liquids where pressure is directly tuned at very low temperature. The other weak point of many experiments is that the pressure is often clamped at room temperature (RT) and thus the tuning through a QCP is achieved through a long discrete process which implies successive coolings from RT. That pushed us in Grenoble to develop a diamond anvil where the pressure medium can be the soft matter He or Ar and due to the small size of the experimental chamber (diameter ≤ 1 mm) a moderate applied strength is sufficient to change in situ the pressure at low temperature ($T \sim 1$ K) up to 10 GPa. The modulation of the strength is simply achieved by changing the ^{4}He pressure in a bellow similarly to the Pomeranchuck cooling of ^{3}He.[40] The pressure is measured in situ by the ruby florescence using an optical fibre. The major experimental achievement is the feedthorough of the electrical leads through the metallic gasket of the pressure chamber. That allows of course to perform four leads resistivity measurements and, even more promising, ac calorimetric detection of phase transitions, by using a metallic thermocouple directly soldered on the sample and an ac input power given by a laser diode.[41] This new technique will be soon applied to three different cases: the QCP of $CeRu_2Ge_2$, the coexistence of magnetism and superconductivity of $CePd_2Si_2$ and the location of P_Δ, P_c the gap closing and of the QCP in SmS. Let us summarise the interest of such studies.

4.1 The case of $CeRu_2Ge_2$: localisation of the f electron, Fermi surface magnetic ordering and QCP

At $P = 0$, $CeRu_2Ge_2$ has a ferromagnetic ground state;[42] its Fermi surface corresponds to a picture where the 4 f electron is treated as localised (small Fermi surface).[22,23] For a pressure $P \sim 8$ GPa, as the volume shrinks, one recovers the $CeRu_2Si_2$ situation at $P = 0$ GPa where the Fermi surface is large as the 4f electron must be considered now as itinerant[21] (Fig. 2). As expected from the experiments made on the $CeRu_2(Ge_{1-x}Si_x)_2$ family by doping, the $CeRu_2Ge_2$ QCP is at $P_c \sim 8$ GPa.[43–45] Furthermore, another localisation of the 4f electron seems to occur by polarising $CeRu_2Si_2$ above its so-called metamagnetic transition at $H_M \sim 7.4$ T at $P = 0$.[46] In the polarised phase ($H \geq H_M$), the Fermi surface topology measured so far is rather near to that found in $CeRu_2Ge_2$ at $P = 0$ however some heavy mass portions are still missing since the integration of the published Fermi surface sheets with their effective masses give a too low contribution for γ.[21,46] The naive idea is that the minority spin becomes more and more heavy on travelling from site to site and their huge effective masses may prevent the detection of their orbit. The particular interesting feature of $CeRu_2Ge_2$ is that at $P = 0$, the AF phase with ordered wavevector $\mathbf{Q}$ similar to that found in $Ce_{1-x}La_xRu_2Si_2$ or $CeRu_2(Ge_{1-x}Si_x)_2$ succeeds to the PA state but a F ground state is well established at lower temperature.[42] Measurements of the specific heat, well below T_C, show a low residual value of γ which agrees with the picture of the 4f localisation and, just below T_N, a large value of C/T weakly temperature dependent which seems to be extrapolated to a large γ term at $T \to 0$ K near that found $\gamma_c \sim 700$ mole$^{-1K^{-2}}$ in the La alloy family near their QCP (Fig. 3). Increasing the pressure up to P^*, T_C collapse, the ferromagnetism disappears, the only ordered phase is AF and the QCP will be achieved for $P_c = 8$ GPa. To summarise, the beauty of $CeRu_2Ge_2$ is that the magnetic phases are certainly associated with a drastic modification of the Fermi surface. One can speculate an expansion of the Fermi surface at P^* through a first order transition as it occurs at the AF-F boundary. At $P = 0$, the microscopic magnetic behaviour has been studied in depth by neutron scattering experiments on $CeRu_2Ge_2$ and $CeRu_2Si_2$ with for the latter a continuous tuning through the metamagnetic transitions.[30] Experiments up to the QCP have been performed by many authors under P in $CeRu_2Ge_2$ with steatite transmitter medium.[43–45] Recently specific heat measurements have been achieved in Geneva[47] in similar conditions. Identical experiments have been made now in Grenoble using the new technology (Fig. 4): hydrostatic He medium and continuous P sweep at low temperature. Focus must be given on the C anomalies at T_C and T_N. The striking point is the rounding of the specific heat anomaly by comparison with the expected molecular field behaviour as $T_N \sim 0$.[34] There is yet no understanding for this broadening; further experiments will be realised below 1 K.

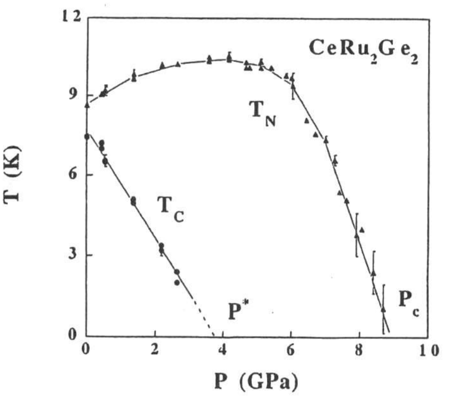

Fig. 2. T, P phase diagram of $CeRu_2Ge_2$ after reference (43). The second order transition line separates the paramagnetic (PA) and antiferromagnetic (AF) phases; the first order transition line is between the AF and ferromagnetic phases. At $T = 0$, the respective characteristic pressures are P_c and P^*. similar results were obtained in the reference (44, 45).

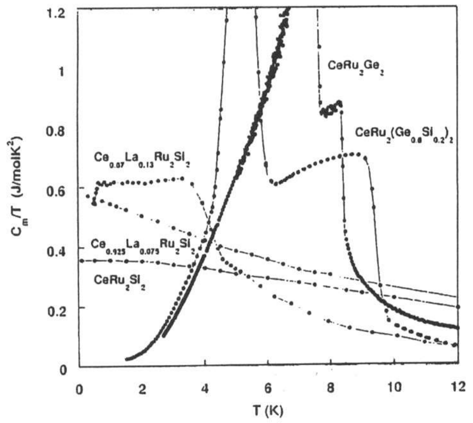

Fig. 3. Temperature variation of C/T (from reference (42)) measured in $CeRu_2Ge_2$ at $P = 0$ and data obtained on alloying systems $CeRu_2(Ge_{1-x}Si_x)_2$ and $Ce_{1-x}La_xRu_2Si_2$.

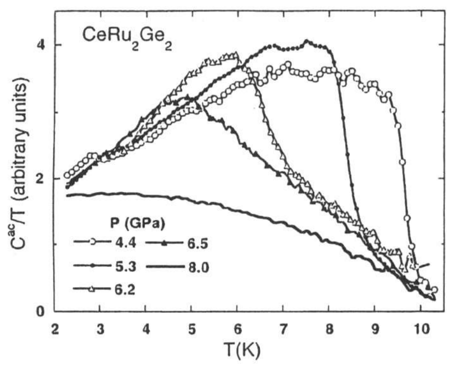

Fig. 4. Specific heat anomaly of $CeRu_2Ge_2$ measured below the QCP using ac calorimetry and the new clean pressure system described in the text.

4.2 *Coexistence of magnetism and superconductivity: case of $CePd_2Si_2$*

The first emergence of superconductivity near a QPC was demonstrated on $CeCu_2Ge_2$ for $P_c = 6$ GPa.[48] Recently clear evidence was found on $CePd_2Si_2$ and $CeIn_3$.[49] The coexistence of superconductivity and AF in heavy fermion systems is not necessary coupled with the proximity of a QCP; a counter example is UPd_2Al_3.[50] The freedom of the parameters is large and a general statement can be irrelevant. However it is clear that now there is a large variety of cases where superconductivity appears just below the QCP; that supports strongly a magnetic origin of the Cooper pairing. Recent experiments realised in Grenoble in a pressure cell with an organic liquid medium on a high quality crystal of $CePd_2Si_2$ obtained in Geneva confirm the previous Cambridge results: emergence of superconductivity just below the QCP (Fig. 5). T_c, the superconducting transition temperature, at the QCP ($P_c \sim 2.6$ GPa) is 395 mK, the resistive width of the superconducting transition is 20 mK. A 7 kbar decrease from the QCP gives a $T_c \sim 180$ mK; the transition width is so large that the resistivity drop is incomplete (Fig. 6). The comparison of these results with data obtained previously in steatite pressure medium in Geneva on a sample of the same batch[51] together with the correlation between the visibility of the resistivity anomalies at T_N or T_C and the pressure dependence of these characteristic temperatures point out the key role of the homogeneity of both the material and the pressure.[52] Concerning the behaviour of the normal phase near a QCP, the main feature is the drastic change of the temperature dependence of the resistivity on each side. Just below P_c, at 24.5 kbar, the low temperature resistivity can be analysed by a sum of T^2 Fermi liquid term plus a spin wave contribution while at $P_c \sim 26$ kbar, the $T^{1.3}$ law is confirmed (Fig. 7). This behaviour is observed up to 40 K. In the near future, continuous clean pressure experiments will be performed with the aim to tune continuously through P_c (persistence of the T^2 resistivity law in AF state , calorimetric proof of the coexistence of AF and supraconductivity and full determination of T_N and T_c boundaries) and to extend the measurements up to 10 GPa ie far above P_c.

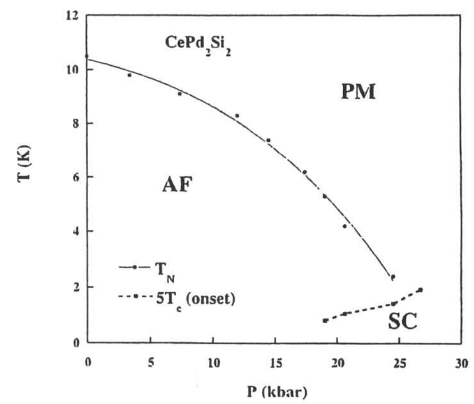

Fig. 5. (T, P) phase diagram of $CePd_2Si_2$. The full line describes the AF - PA boundary, the dashed line shows the superconducting (SC) boundary. For clarity, the T_c value has been multiplied by 5.

4.3 *Insulator in intermediate valent phase. Gap closing and QCP: case of SmS*

The new pressure facility notably to sweep continuously the pressure at very low temperature ($T \leq 1K$) will lead to revisit an unconventionnal phase diagram: SmS. Three decades ago[53] an important discovery was the isostructural first order transition of SmS characterised by a $T_{B \rightarrow G}$ line (Fig. 8) between a low pressure black (B) insulating phase where the Sm is divalent and a gold phase (G) where the Sm is clearly in an intermediate valence state ($\vartheta \sim 2.7$). In the G phase, the particular interest is that the light 5d electronic carrier is released by the valence mixing according to the equilibrium:

$$Sm^{2+} \Longleftrightarrow Sm^{3+} + 5d.$$

The electrical resistivity at room temperature is that of a bad metal (~ 300 $\mu\Omega cm$) just above $P_{B \rightarrow G} \sim 7$

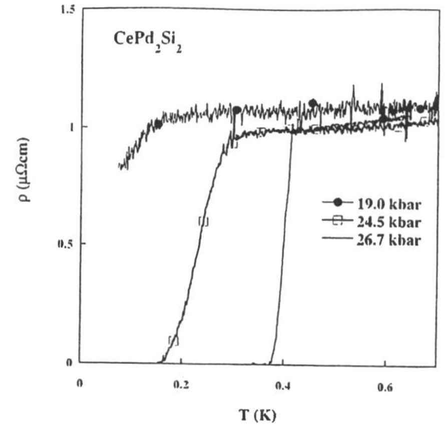

Fig. 6. Resistivity superconducting drops of $CePd_2Si_2$ below the QCP for three different pressures.

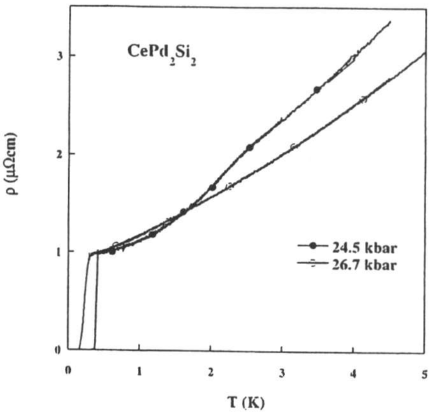

Fig. 7. Temperature dependence of the resistivity ρ just below the QCP and at the QCP.

GPa. Just near $P_{B\to G}$ a continuous increase of ρ is observed on cooling; in its low pressure range the gold phase ends up below a crossover temperature T_Δ in an insulating ground state as also observed for SmB_6, YbB_{12} or TmSe.[19] In all pressure resistivity experiments, above a well defined critical pressure $P_\Delta = 2$ GPa, the ground state is metallic.[54–56] By further increasing the pressure, a magnetic QCP will be reached at P_c when the Sm ion approaches its trivalent state of a Kramer ion. The two goals in a new generation of experiments will be to determine the intersection of $T_{B\to G}$ and T_Δ at very low temperature (interest of continuous tuning) to precise in the same temperature range the location of P_Δ and P_c and, if excellent samples become available, to search for superconductivity.

§5. Conclusion

Our aim here was to stress the importance of new experimental approaches. A new generation of experiments is needed to get a deeper insight into heavy fermion systems. Recent pressure developments in instrumentation (hydrostaticity, continuous tuning), open the possibility to explore in clean extreme conditions (P, H, T) the

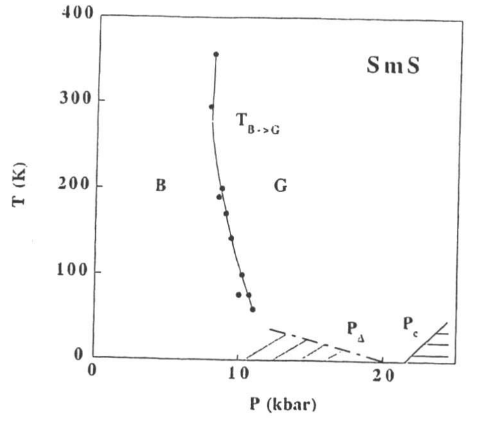

Fig. 8. Schematic (T, P) phase diagram of SmS. The full line $T_{B\to G}$ is the first order transition line between the black and the gold phases, the dotted-dashed line mimics the pressure dependence of T_Δ the crossover metal-insulator temperature, $T_\Delta = 0$ for $P = P_\Delta$. Here it is suspected that $P_\Delta \leq P_c$.

QCP, the interplay of magnetism (PA, AF, F phases) and metallic conduction (superconductivity, metal insulator transition, role of disorder). Calorimetric measurements under pressure open new perspectives as they are simple to interpret than transport measurements where the scattering time must be considered. It is crucial to have also microscopic probes. Few elastic neutron scattering experiments have been performed under pressure; recent progress has been made by NMR at least up to 25 kbar.[57,58] Open perspectives are studies of magnetic scattering using synchrotron facilities and any technique able to identify the defects in real space and the concomitant electronic properties in the surrounding (scanning tunnelling microscope, ...). Progress in the field is directly related to the improvement of the material. In new systems like $CeIn_3$, $CePd_2Si_2$ and $CeNi_2Ge_2$ excellent single crystals can be produced ($\rho_0 \leq 1\mu\Omega cm$), in contrast to the case of $CeCu_2Si_2$ which leads to a poor knowledge of its magnetism.[59,60]

The complete determination of the SmS phase diagram is a good example of an open problem which could be solved completely if a major advance is made now in the material.

One of us (J. Flouquet) likes to thank deeply Pr. Y. Miyako for his continuous support to facilitate our collaboration with the Japanese physicists. In the last decade it was a pleasure to have a strong and fair competition with his team. His recent successes in the studies of $Ce(Ru, Rh)_2Si_2$ and URu_2Si_2 are excellent examples of experiments or ideas in which we have failed or missed.

Note added: The second low temperature antiferromagnetic transition at T_L (L means locking) of $CeRu_2Ge_2$ under P is not discussed for simplicity in this paper.

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