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Interplay and competition in heavy-fermion systems

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Heavy-fermion systems offer unequalled opportunities to study a wide variety of physical phenomena, such as the Kondo (lattice) effect, the RKKY interaction, (weak) long-range (anti)ferromagnetic order, metamagnetism and unconventional (non-singlet) superconductivity. In the past few years a broad experimental and theoretical expertise has been applied in an attempt to unravel these phenomena and their interplay. Three main issues have emerged: the competition between the Kondo and RKKY interaction, the proximity of a magnetic instability and the interplay of superconductivity and the antiferromagnetic interactions. We review the recent developments in heavy-fermion physics based on these main issues.

1. Introduction

The discovery of the formation of a highly correlated electron band at the Fermi level at low temperatures in some Ce, Yb and U intermetallic compounds has given rise to a vast research area, now commonly referred to as heavy-fermion physics [1]. The origin of the heavy-fermion behaviour lies in a wealth of electronic and magnetic interactions that are brought about by a delicate degree of delocalization of the 4f or 5f electrons. The degree of itineracy of the f electrons depends on the complex hybridization with the p or d orbitals of the ligand atoms. The hybridization is extremely sensitive to shape and volume effects, and moderate external or chemical pressures can easily change the low-temperature density of states by a factor of 10 to 100. Concurrently, the characteristic energies of the interactions that build up the heavy fermions can be tuned to a large extent, and a wide range of phenomena is observed: the Kondo (lattice) effect, the RKKY interaction, (weak) long-range (anti)ferromagnetic order, pseudo-metamagnetism and unconventional (non-singlet) superconductivity. In the past few years much experimental and theoretical work has been devoted to a study of these phenomena and their interplay. Three main issues have emerged: (i) the competition between the Kondo and the Ruderman–Kittel–Kasuya–Yosida (RKKY) interaction, (ii) the proximity of a magnetic instability and (iii) the interplay of superconductivity and the antiferromagnetic interactions. In this paper we illustrate these main issues by a number of experimental studies on exemplary systems, like U(Pt, Pd)$_3$ and (Ce, La)Ru$_2$Si$_2$.

2. Kondo versus RKKY interaction

Evidence for competing electronic interactions in heavy-fermion systems has for the greater part been gathered by alloying studies, i.e. by progressive replacements of one of the constituents. Detailed measurements of the transport, magnetic and thermodynamic properties along such a series often yield distinctly different regimes. Let us illustrate this with the pseudobinary compound U(Pt$_{1-x}$Pd$_x$)$_3$ [2]. In pure UPt$_3$ the low-temperature properties are dominated by antiferromagnetic interactions, while by substituting small amounts of Pt by isoelectronic Pd, a crossover to a regime dominated by Kondo properties is observed. This change in regime is most clearly demonstrated by the electrical resistivity, $\rho(T)$ (see fig. 1). For pure UPt$_3$ the gradual drop of $\rho(T)$ with decreasing temperature is ascribed to the stabilization of the antiferromagnetic correlations, while for a Pd content of only 10 at% a Kondo-like upturn is observed. The maximum in the susceptibility, $\chi(T)$, at $T_{\max} = 18$ K, and the metamagnetic-like transition at a field of 20 T ($T \leq T_{\max}$),

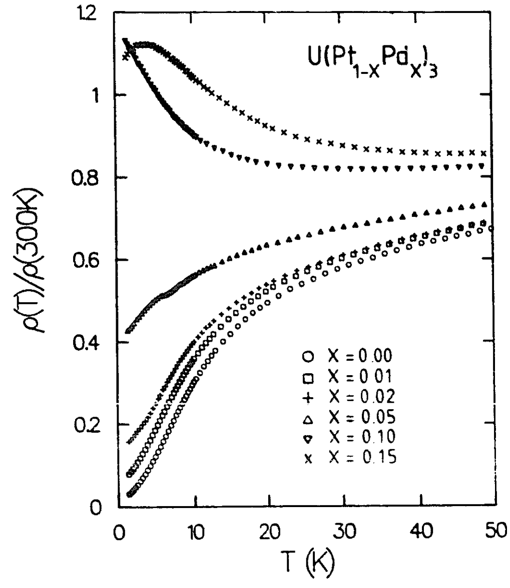

Fig. 1. Electrical resistivity of polycrystalline U(Pt$_{1-x}$Pd$_x$)$_3$ for $x \leq 0.15$. The resistance values have been normalized to one at $T = 300$ K. After ref. [2].

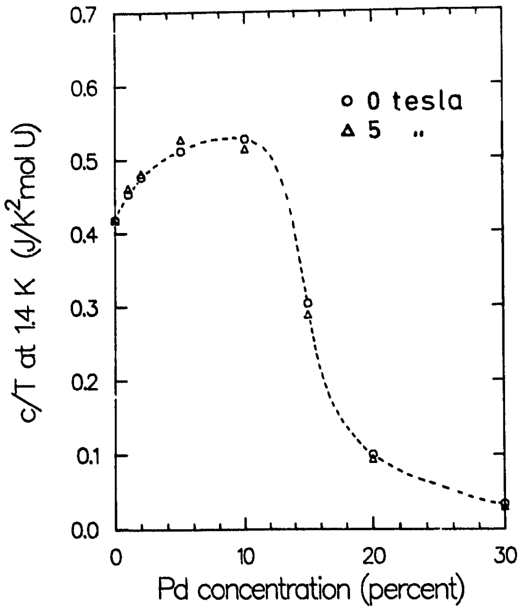

Fig. 2. The c/T value at $T = 1.4$ K for polycrystalline $U(Pt_{1-x}Pd_x)_3$ for $x \leq 0.30$, in zero field (o) and in a field of 5 T (Δ). Note the changes in sign of the field effect near $x = 0.10$. The dashed line serves as a guide to the eye. After ref. [2].

characteristic of pure UPt_3, gradually decrease on alloying and are no longer observed for $x = 0.10$, lending further support for a suppression of the antiferromagnetic correlations. Consistent herewith it is observed that the linear coefficient of the electronic specific heat (γ) passes through a pronounced maximum near $x = 0.10$ (fig. 2), while $\partial\gamma/\partial B$ changes sign between $x = 0.07$ and $x = 0.10$, (as $\partial\rho/\partial B$ does at low temperatures). In the same Pd concentration range the coefficient of volume expansion ($\alpha_v(T)$) changes sign and the Grüneisen parameter, $\Gamma(T \rightarrow 0)$, shows a huge drop from 75 for pure UPt_3 to -300 for $x = 0.15$ [3]. This salient change in regime clearly indicates the presence of competing electronic interactions and is more generally attributed to a competition between the RKKY (T_{RKKY}) and Kondo effect (T_K) [4]. However, the variation of T_{RKKY} and T_K with Pd content is not easily determined as both mechanisms are present [5]. Besides part of the fluctuating moment orders antiferromagnetically [2] (see section 3), giving rise to yet another superimposed contribution. In order to unravel the composite low-temperature properties of the U(Pt, Pd)$_3$ system, inelastic-neutron scattering experiments will certainly be very helpful. Inelastic neutron scattering experiments on pure UPt_3 [6] have warranted a description in terms of the competing electronic interactions. In this respect it is interesting to compare UPt_3 with $CeRu_2Si_2$, which has rather similar transport, magnetic and thermodynamic properties [7]. Inelastic neutron-scattering experiments on $CeRu_2Si_2$ yield two low-temperature contributions: on-site interactions (Kondo) and intersite interactions (antiferromagnetic correlations). At the metamagnetic-like transition ($B^* = 8$ T for the pure compound) the intersite contribution is drastically reduced, as follows from neutron-scattering data in the field region [8]. Another way to get more insight into the Kondo contribution is by performing measurements in the field region where the intersite contribution is largely suppressed ($B \gg B^*$). This requires, however, very strong magnetic fields. In the case of $CeRu_2Si_2$ such measurements have been performed recently. Specific-heat measurements up to 20 T [9], in the temperature interval 1.5 $K < T < 30$ K, reveal a pronounced maximum of c/T taken at 1.5 K at B^*. The field enhancement of the effective mass at B^* amounts to $m_{eff}(B^*)/m_{eff}(B = 0) = 1.28$ (see fig. 3). As the linear regime in $c(T)$ close to B^* is only attained at very low temperatures a considerable sharpening of $\gamma(B)$ close to B^* is expected. This was recently con-

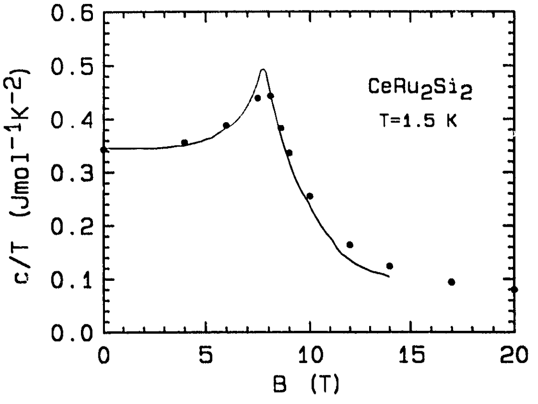

Fig. 3. The measured c/T-value at $T = 1.5$ K as a function of the magnetic field (●) for single-crystalline $CeRu_2Si_2$ (B along the tetragonal axis). The solid line has been calculated from the magnetization data. After ref. [9].

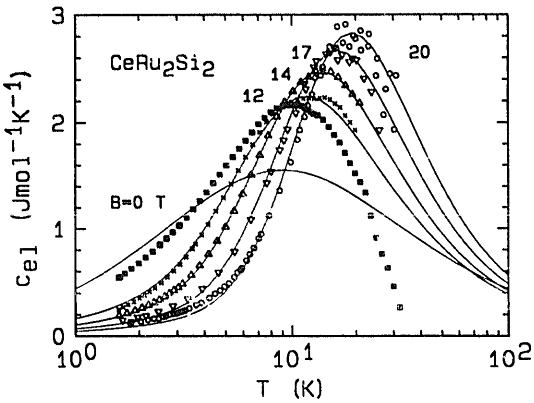

Fig. 4. The electronic specific heat of $CeRu_2Si_2$ for $B = 0$ T (■) and for fields $B > B^*$ along the tetragonal axis on a logarithmic temperature scale. The solid lines are a comparison with a single-resonance level model. After ref. [9].

firmed by low-temperature ($T > 0.1$ K) magnetization measurements, from which it has been inferred that $m_{\mathrm{eff}}(B^*)/m_{\mathrm{eff}}(B=0) = 1.77$ for $T \to 0$ [10]. In the high-field region ($B > B^*$) the Kondo contribution remains. It is gradually suppressed with increasing field (see fig. 4). The reduction of the effective mass (in 20 T) amounts to about 80% with respect to the zero-field value ($\gamma(B=0) = 350$ mJ/mol K^2). In the high-field region ($B \geq 12$ T) a reasonable fit to the data can be made with a single-resonance level model (see fig. 4).

In many other compounds with an enhanced effective mass the competition of the RKKY and the Kondo effect plays a major role, although its effect on the low-temperature properties is often less dominant (see, e.g., $CeCu_6$ [8] and $CePt_2Si_2$ [11]).

3. The magnetic instability

An implicit consequence of the competition between the RKKY and the Kondo effect is the proximity of a magnetic instability [4]. Indeed by small changes in the chemical composition, long-range magnetic order with fairly large magnetic moments ($|\mu| \approx 0.5\mu_B$/f atom) can be induced in most of the heavy-fermion compounds. For instance, in the case of $Ce_{1-x}La_xRu_2Si_2$ [12] long-range antiferromagnetic order is found for $x \geq 0.07$, and in the case of $CeCu_{6-x}Au_x$ long-range order occurs for $x \geq 0.1$ [13]. The approach of the magnetic instability is explained by the increase in volume that is induced by alloying. In general, the volume increase will lead to a weaker hybridization and thus to a reduction of the exchange coupling constant J, i.e. an increase in the local character of the f electrons. As $T_{\mathrm{RKKY}} \propto J^2$ and $T_{\mathrm{K}} \propto e^{-1/|J|}$, the RKKY interaction dominates at small J and (anti)ferromagnetic order will occur. The strong volume dependence of the characteristic energies in heavy-fermion compounds is furthermore confirmed by the unusually large low-temperature Grüneisen parameters, i.e. 100 times larger than in ordinary metals [14]. Also the volume increase when entering the antiferromagnetic state [15] and the (in general) negative hydrostatic pressure effect on T_N are consistent with the increase in local character of the f electrons. However, as most heavy-fermion systems have a strongly anisotropic hybridization, J is obviously not always governed by the volume effect. For instance, in the $U(Pt_{1-x}Pd_x)_3$ series long-range antiferromagnetic order, with a maximum Néel temperature (T_N) of 5.8 K, has been found in the concentration range $0.02 \leq x \leq 0.07$ [2]. As Pd is smaller than Pt the volume decreases; however, the tendency of the f moments to localize is here probably related to a subtle change in the c/a ratio of the hexagonal lattice. An important observation that can be made for many of the heavy-fermion alloys is that only a part of the fluctuating moment orders, that is the heavy-fermion properties survive in the ordered state. Consequently the entropy involved in the ordering is small. While the magnetic instability is easily attained in the Pauli-paramagnetic heavy-fermion systems, the reverse is observed in the antiferromagnetically ordered systems. For instance, in the case of $CePd_2Si_2$ ($T_N = 9$ K) both Y and La doping lead to a decrease of the ordering temperature (T_N of $CePd_2Si_2$ is just at the maximum of the $T_N(J)$ curve in Doniach's phase diagram) [16]. In URu_2Si_2 ($T_N = 17.5$ K) both Rh [17] and Tc [18] doping lead to a suppression of T_N. Obviously, the volume is here not the dominant parameter. Dilution effects and anisotropic hybridization must be considered in any attempt to model the observed variations of T_N.

A hitherto unexplained and puzzling property of heavy-fermion compounds is the so-called small-moment magnetism. Microscopic techniques, like μSr, NMR and neutron scattering have revealed that the heavy-fermion compounds URu_2Si_2, $CeAl_3$ ($T_N \leq 1$ K), UPt_3 ($T_N = 5$ K) and (superconducting) $CeCu_2Si_2$ ($T_N = 0.6$ K) exhibit weak antiferromagnetic order with in some cases very small moments, i.e. in the order of $0.01\mu_B$/f-atom (see ref. [19] and references therein). Among the prominent heavy-fermion compounds, apparently only $CeRu_2Si_2$ and $CeCu_6$ remain in a Pauli paramagnetic ground state. Recently it was reported that superconducting UBe_{13} ($T_c = 0.9$ K) also orders antiferromagnetically as evidenced by a magnetostriction technique ($T_N = 8.8$ K) [20]; however, this could not be confirmed by our recent data [21]. The small-moment magnetism in $CeAl_2$, $CeCu_2Si_2$ and UPt_3 has thus far not been probed convincingly by means of a macroscopic technique, like the specific heat, which makes a detailed study difficult. The connection between the small-moment and the large-moment magnetism (i.e. the moment induced by alloying) is by no means clear. Therefore, it would be interesting to study the evolution of the tiny moment on alloying, in particular in the case that this leads to a large moment state, as for instance in the series U(Pt, Pd)$_3$. Finally, we remark that at present it cannot be completely excluded that the emergence of the tiny moment is related to metallurgical problems (defects etc.).

4. Interplay of superconductivity and antiferromagnetism

The occurrence of superconductivity in materials where strong electronic interactions build up the heavy quasiparticles prompted questions about unconventional superconductivity. As it are the heavy quasiparticles that form the superconducting condensate and as the uppercritical field is unusually large for superconductors with such a low value of T_c, it was proposed that the superconducting Cooper pairs are mediated by an electron–electron interaction, in particular by antiferromagnetic correlations, instead of by the usual

Table 1
Grüneisen parameters for the superconducting transition temperature $\Gamma_{T_c} = -\partial \ln T_c/\partial \ln V$ and for the characteristic temperature of the Fermi liquid $\Gamma_{T^*} = -\partial \ln T^*/\partial \ln V$, for UPt_3, URu_2Si_2 and UBe_{13}. All values have been determined from specific heat measurements under pressure [23,24]

	UPt_3	URu_2Si_2	UBe_{13}
Γ_{T_c}	−65	−59	−21
Γ_{T^*}	+50	+40	+52

electron–phonon interaction. Evidence for unusual superconductivity is provided by the strongly anisotropic gap function as evidenced by the observation of power laws for the temperature dependence of the electronic excitation spectrum below T_c. Besides, in some cases unusual anomalies in the upper critical field, $B_{c2}(T)$, and additional anomalies within the superconducting phase have been observed. An important question, at present under investigation, is whether the double superconducting transition in UPt_3 [22] is caused by a coupling of the superconducting and antiferromagnetic (i.e. the small moment magnetism observed below $T_N = 5$ K) order parameters. If this turns out to be so, some preliminary evidence for the interplay of magnetism and superconductivity in the U-based heavy-fermion superconductors is provided. However, a definite proof that electron–electron interactions are the microscopic basis for heavy-fermion superconductivity is still not available. In an attempt to couple the characteristic energy for the electronic interactions to the superconducting transition temperature, we can compare their relative volume dependences or Grüneisen parameters, by defining $\Gamma_{T_c} = -\partial \ln T_c/\partial \ln V$ and $\Gamma_{T^*} = -\partial \ln T^*/\partial \ln V$, where $T^* \propto T_F \propto \gamma^{-1}$ and T_F is the renormalized Fermi temperature. In table 1 we compare values for Γ_{T_c} and Γ_{T^*} for UBe_{13}, UPt_3 and URu_2Si_2 determined in one single experiment on one sample: specific heat measurements under pressure [23,24] that probe both T_c and the normal state γ. Similar Grüneisen parameters have been determined from resistivity and susceptibility measurements under pressure and from thermal expansion measurements [14]. However, due to sample-dependent properties a rather large spread in the reported values is found. We conclude that for all three systems Γ_{T_c} and Γ_{T^*} are of the same order of magnitude with opposite sign. The inverse correlation of Γ_{T_c} and Γ_{T^*} justifies a further investigation of Cooper pairing mediated by antiferromagnetic interactions.

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