Neutron Scattering Studies of Heavy Fermion Systems

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Abstract

We review the results of recent studies of the elastic and inelastic neutron scattering from a variety of heavy fermion compounds. This class of materials exhibits a rich variety of ground states: antiferromagnetically ordered, superconducting, semiconducting, and paramagnetic. Neutron scattering from single crystals and powders has been a productive tool for probing the magnetic order and fluctuations in all four cases. This review deals with work on UPt_3, UPd_2Al_3, UNi_2Al_3, UNi_4B, CeNiSn, $Y_{1-x}U_xPd_3$, and $UCu_{5-x}Pd_x$.

1 Introduction

1.1 Overview

Heavy fermion compounds, typically alloys containing U or Ce, are characterised by the small energy scale associated with the hybridization of nearly localised *f*-electrons with conduction electrons. This small energy scale means that properties such as band structure, which are normally not considered temperature dependent, can vary with temperature and are sensitive to small perturbations. This sensitivity gives rise to a rich variety of low temperature states in these materials; for a review see Grewe and Steglich (1991).

At high temperatures heavy fermion systems behave as Kondo lattices and the unpaired *f*-electrons have a local magnetic moment that interacts with the conduction electrons in the same way as an isolated Kondo impurity in a metal. As the temperature is lowered, however, the magnetic moments no longer behave as isolated, localised impurities and the system enters the coherent state which is characterised by the large effective mass (and enhanced electronic specific heat) associated with the quasiparticles of a strongly interacting band of carriers. In this coherent, heavy fermion state there are substantial antiferromagnetic spin fluctuations which can be studied in great detail by magnetic neutron scattering from single crystals. This has been the topic of a recent review (Aeppli and Broholm, 1994). The present paper presents highlights of some experiments which have occurred since then.

1.2 Neutron scattering cross section

Because of its magnetic moment the neutron can couple to moments in solids via the dipolar force. The energy and wavelengths of thermal and subthermal neutrons are well matched to the energy and length scales of most condensed matter systems and this is particularly true for heavy fermions. We will briefly review the formalism which describes the magnetic neutron scattering. For a detailed treatment of the neutron scattering cross-section there are some excellent texts which can serve as an introduction (Squires, 1978) or more comprehensive exposition (Lovesey, 1984).

The partial differential cross section for magnetic neutron scattering, which measures the probability of scattering per solid angle per unit energy, is

$$\frac{d^2\sigma}{d\Omega dE'} = \frac{k'}{k}\frac{N}{\hbar}(\gamma r_o)^2 \left|f(\boldsymbol{Q})\right|^2 \sum_{\alpha\beta}(\delta_{\alpha\beta} - \hat{Q}_\alpha \hat{Q}_\beta) S^{\alpha\beta}(\boldsymbol{Q},\omega) \qquad (1)$$

where $k(k')$ is the incident (scattered) neutron wavevector, N is the number of moments, $\gamma r_o = 5.391$ fm is the magnetic scattering length, $f(\boldsymbol{Q})$ is the magnetic form factor (analogous to the electronic form factor appearing in the x-ray scattering cross section), $\boldsymbol{Q}$ is the momentum transfer, ω is the energy transfer, and the summation runs over the Cartesian directions. $S^{\alpha\beta}(\boldsymbol{Q},\omega)$ is the magnetic scattering function which is proportional to the space and time Fourier transform of the spin-spin correlation function.

If the incident and scattered neutron energies are the same (elastic scattering) then the correlations at infinite time are being probed and, in a magnetically ordered material, the scattering function will contain delta functions at the wavevectors corresponding to magnetic Bragg reflections. The $(\delta_{\alpha\beta} - \hat{Q}_\alpha \hat{Q}_\beta)$ term in the cross section means that neutrons probe the components of spin perpendicular to the momentum transfer, $\boldsymbol{Q}$. If there is no analysis of the scattered neutron energy then (within the static approximation) the measured intensity is proportional to the Fourier transform of the instantaneous correlation function which is essentially a snapshot of the spin correlations in reciprocal space. At non-zero energy transfers the spin dynamics of the system under study are probed. In a magnetically ordered system of localised spins the elementary magnetic excitations are spin waves.

The fluctuation dissipation theorem relates the correlations to absorption, in other words the scattering function is proportional to the imaginary part of a generalised ($\boldsymbol{Q}$ and ω dependent) susceptibility, $\chi''(\boldsymbol{Q},\omega)$. In the zero frequency, zero wavevector limit, the real part of the generalised susceptibility is the usual DC susceptibility measured by magnetisation. In a metal the elementary excitations are electron-hole pairs. Since it is possible to excite an electron-hole pair by promoting a quasiparticle from below the Fermi surface to above the Fermi surface, and at the same time flipping its spin, neutrons can be used to probe the low energy excitations of a metal. The generalised susceptibility (for a non-interacting metal) is just the Lindhard susceptibility which can be calculated from the band structure.

2 Antiferromagnetism and superconductivity

2.1 $\mathrm{UPt_3}$

$\mathrm{UPt_3}$ has remained a very popular system because it is both the quintessential strongly renormalized Fermi liquid, as revealed especially by de Haas–van Alphen experiments, and the quintessential unconventional superconductor, displaying an array of properties ranging from multiple superconducting phases to anisotropies not likely predicated on normal state anisotropies. While the broad outlines of the $\mathrm{UPt_3}$ problem were clear several years ago, the past two years have witnessed scattering experiments which have answered important outstanding questions. These experiments all have to do with the weak antiferromagnetic order whose Bragg signal is reduced by passing into the superconducting state, and which is greatly enhanced – while superconductivity is eliminated – upon Th substitution for U or Pd substitution for Pt (Ramirez et al., 1986; de Visser et al., 1986; Goldman et al., 1987; Frings et al., 1987). In particular, Isaacs et al. (1995) performed a combination of x-ray and neutron diffraction experiments which showed the following (see Fig. 1): (i) The reduction in the magnetic Bragg scattering found in an earlier experiment (Aeppli et al., 1989) is due to a reduction in the magnitude of the corresponding magnetic moment, and not to rotation of the moments, e.g., in the basal planes of the material. (ii) There seems to be little difference between the behaviours of the magnetic order exhibited in the near surface region probed by resonant x-ray scattering and the bulk probed by neutrons. (iii) The magnetic coherence length in quite heavily doped and non-superconducting $\mathrm{U_{0.95}Th_{0.05}Pt_3}$ is resolution-limited. This again emphasizes that a special local disorder is the most likely cause of the magnetism in pure $\mathrm{UPt_3}$.

The second new scattering experiment also addressed the vector nature of the ordered moment. In particular, Lussier et al. (1996) investigated whether an external magnetic field parallel to the basal planes – the 'easy' direction as inferred

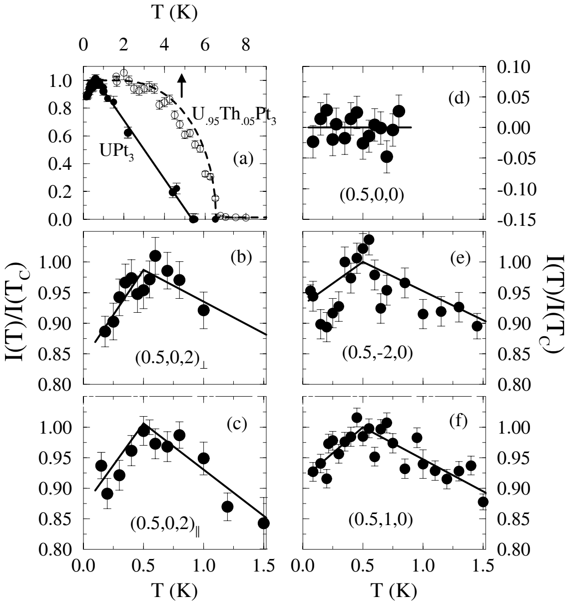

Figure 1. Temperature dependence of the antiferromagnetic Bragg peaks for UPt_3. (a)–(c) show the intensity measured with x-rays (with neutron data for isostructural $U_{0.95}Th_{0.05}Pt_3$ shown in (a) [open circles] for comparison). (d)–(f) show the neutron scattering intensity for three different Bragg reflection entering the superconducting state. From Isaacs et al. (1995).

from bulk measurements – could rotate the moments. A field of up to 3.2 T was not able to either rotate the moments or select a single domain (see Fig. 2). Given that such a limiting field is beyond H_{c2} for the superconductor, finding (i) of Isaacs et al. (1995) is not surprising. Thus there are anisotropies, possibly random, which strongly pin the small ordered moment in pure UPt_3. It will be interesting to see whether the same result is obtained in the more coherent antiferromagnetic state induced by Th and Pd impurities. The finding that a single magnetic domain is not produced implies that either the magnetic structure is not single-$\boldsymbol{Q}$ or all measurements of the superconducting phase diagram have been in multi domain samples, requiring a re-evaluation of theories based on the symmetry breaking of antiferromagnetic ordering.

2.2 UPd_2Al_3 and UNi_2Al_3

In 1991 two new heavy fermion compounds were discovered which displayed the coexistence of antiferromagnetic order and superconductivity. UPd_2Al_3 has an

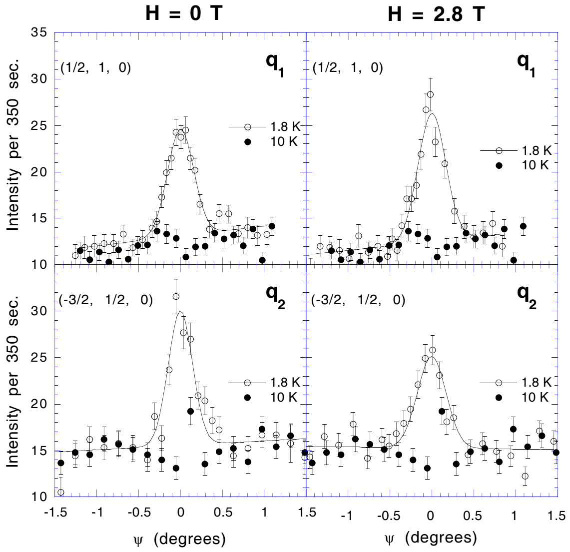

Figure 2. Magnetic Bragg peaks for two different domains in UPt_3 for $H = 0$ and 2.8 tesla. Complete selection of a single domain by the 2.8 T field would eliminate the $\mathbf{q}_2$ Bragg peak and increase the $\mathbf{q}_1$ peak by a factor of three. From Lussier et al. (1996).

antiferromagnetic transition at 14.4 K and, in the best samples, a superconducting T_c of 2 K, the highest of any heavy fermion compound at ambient pressure (Geibel et al., 1991a). UNi_2Al_3 has a somewhat lower T_N (5.2 K) and T_c (1 K) (Geibel et al., 1991b). Both share the same hexagonal crystal structure (space group P6/mmm).

Powder neutron diffraction has shown that, in the antiferromagnetic state, UPd_2Al_3 has moments of 0.85 μ_B lying in the hexagonal basal plane with the moments in a given layer ferromagnetically aligned and alternating up the c axis (Krimmel et al., 1992a). Initial reports of a suppression of the ordered moment in the superconducting state by Krimmel et al. (1992a) have not been reproduced (Kita et al., 1994). Measurements of the magnetisation density in the paramagnetic state using polarized neutrons have shown that the magnetic moment resides on the U site with no spin transfer to the Pd ions (Paolasini et al., 1993), comparison with magnetisation data suggest that there is an additional (12% contribution) from the polarisation of the conduction electrons. A determination of the magnetic phase diagram up to 5 T (Kita et al., 1994) has shown that the moment lies along the a axis in the basal plane. Application of a magnetic field perpendicular to one of the a axes (along $[\bar{1}\,1\,0]$) favours that magnetic domain and as the field is increased

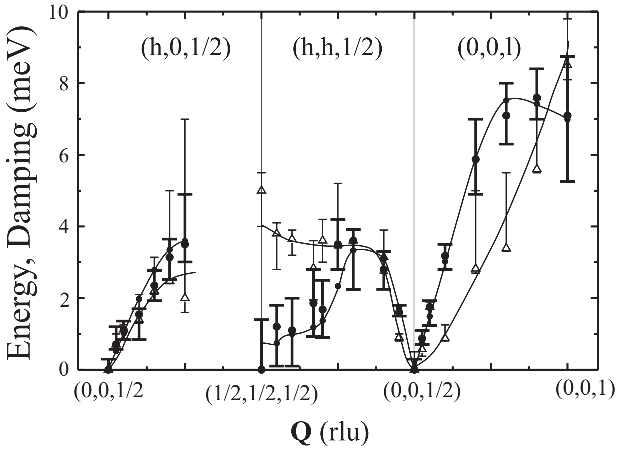

Figure 3. Wavevector dependence of the energy (filled circles) and damping (open triangles) of the spin waves in the ordered state of UPd_2Al_3. There are well defined spin waves along the c^* axis however, in the basal plane, the response is overdamped making it difficult to independently determine $\Gamma_{\boldsymbol{Q}}$ and $\omega_{\boldsymbol{Q}}$.

above a critical field of order 0.5 T the fraction of the sample with moments aligned along the a axis perpendicular to H increases from 33% to 100%. If the field is applied parallel to [0,1,0] then a two step process occurs: first above 0.5 T the two domains at $\pi/3$ are selected, then above 4 T the moments are constrained to lie perpendicular to the field along the next nearest direction in the basal plane. As the temperature is increased towards T_N the fields for domain selection and reorientation approach zero.

The inelastic neutron scattering from UPd_2Al_3 has been studied using powder, time-of-flight (Krimmel et al., 1996) and single crystal, triple-axis techniques (Petersen et al., 1994; Mason et al., 1995). The powder measurements in the paramagnetic state show a strong quasi-elastic response which is peaked at the wavevector corresponding to the $(0, 0, \frac{1}{2})$ Bragg peak. The single crystal studies have shown that in the antiferromagnetically ordered state this response evolves into spin waves which, within the limits of the experimental resolution of 0.35 meV, have no gap at the ordering wavevector. The full dispersion surface extracted from these measurements is shown in Fig. 3 along with the wavevector dependence of the spin wave lifetime. These quantities correspond to the energy and damping of an inelastic Lorentzian response corrected for spectrometer resolution. The structure of the dispersion requires a model of the magnetic interactions with at least four groups of next neighbours implying long range interactions. Moreover, it is not

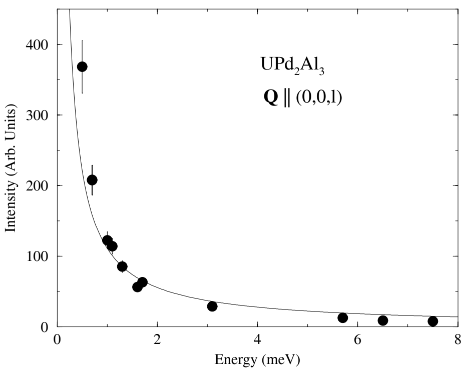

Figure 4. Spin wave intensity as a function of energy for UPd$_2$Al$_3$ obtained for momentum transfers displaced from $(0, 0, \frac{1}{2})$ along the c axis. The intensity is the amplitude for an inelastic Lorentzian response convolved with the spectrometer resolution. The line is the $1/\omega$ dependence expected for conventional spin waves.

possible to describe both energies and lifetimes in a localised moment spin wave model (Lindgård et al., 1967) because damping arising from off-diagonal terms in the Hamiltonian results in a zone centre gap inconsistent with the data. This suggests the damping is of extrinsic (conduction electron) origin. The damping is generally comparable to the spin wave energy although for wavevectors displaced along the c axis there are well resolved modes with a linear dispersion. The intensity of the spin waves along the c axis, obtained from the same fits, is shown in Fig. 4 in comparison with the $1/\omega$ expected for conventional spin waves. Measurements of the spin wave intensities in a single domain sample (produced as described in the preceding paragraph) have shown the excitations are transverse to the moment direction. It appears that UPd$_2$Al$_3$ is unique among U compounds in that it possesses conventional spin wave excitations with a very small or no gap at the ordering wavevector. These spin waves are strongly damped due to interaction with the conduction electrons but, at energies less than a few multiples of $k_B T_c$, show no change on entering the superconducting state (Petersen et al., 1994). This is consistent with the results of heat capacity (Caspary et al., 1993) and muon spin rotation measurements (Feyerherm et al., 1994) which have been interpreted as evidence for two coexisting electronic systems, localised $5f$ magnetic states and delocalised states which are responsible for superconductivity (Steglich et al., 1996).

Initial powder diffraction studies of UNi$_2$Al$_3$ failed to observe any magnetic Bragg peaks below T_N and placed an upper bound on the ordered moment of 0.2

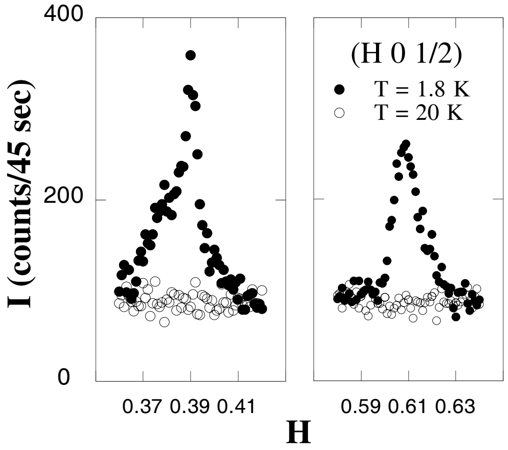

Figure 5. Scans through the incommensurate peaks in $\mathrm{UNi_2Al_3}$ along the $(h, 0, \frac{1}{2})$ direction above (open circles) and below (closed circles) $T_N \sim 5.2$ K. From Schröder et al. (1994)

μ_B (Krimmel et al., 1992b). μSR experiments indicated that the ordered moment was likely of order 0.1 μ_B (Amato et al., 1992). Schröder et al. (1994) performed neutron diffraction measurements on a single crystal of $\mathrm{UNi_2Al_3}$ and found that it ordered incommensurately below 5.2 K with an ordered moment of 0.24 ± 0.1 μ_B. Figure 5 shows scans through two of the incommensurate wavevectors, $(\frac{1}{2} \pm \delta, 0, \frac{1}{2})$ with $\delta = 0.110 \pm 0.003$, above and below T_N. The intensities of six magnetic Bragg peaks measured at 1.8 K are best described by a model structure which is a longitudinal spin density wave within the hexagonal basal plane with the moments parallel to $\mathbf{a}^*$. The moment direction in $\mathrm{UNi_2Al_3}$ is therefore rotated $\pi/6$ compared to $\mathrm{UPd_2Al_3}$ but the observation of an incommensurate modulation within the basal plane is perhaps not surprising given the long range interactions manifested in the spin wave measurements in $\mathrm{UPd_2Al_3}$.

2.3 $\mathrm{UNi_4B}$

One of the most intriguing new compounds which have been studied in recent years is $\mathrm{UNi_4B}$ which has a structure based on the hexagonal $\mathrm{CaCu_5}$ structure. There is a small distortion which modifies the local environment of $\frac{2}{3}$ of the U ions through their collective motion towards the remaining sites (Mentink et al., 1996a). As a result $\frac{1}{3}$ of the uranium moments are on six-fold symmetric sites while the remainder are on two-fold symmetric sites. The resistivity, susceptibility, and

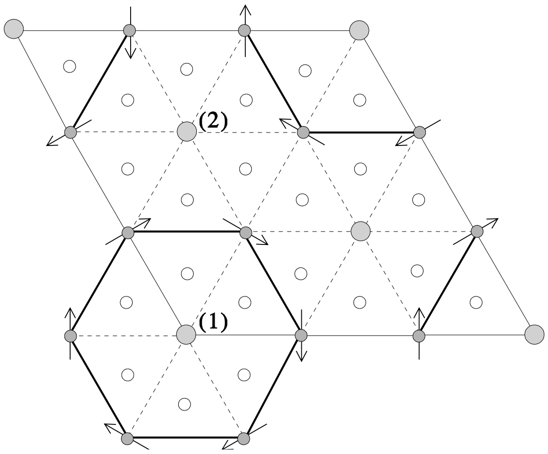

Figure 6. Magnetic structure of UNi_4B projected onto the hexagonal basal plane. The magnetic layers are stacked ferromagnetically along the *c* axis. The solid circles, labelled (1) and (2), represent the paramagnetic U moments. From Mentink et al. (1994).

specific heat of UNi_4B all show anomalies typical for antiferromagnetic ordering at 21 K and this has been confirmed by single crystal neutron diffraction (Mentink et al., 1994). The magnetic structure, shown in Fig. 6, is very unusual. The moments on the outer, two-fold, sites of hexagonal plaquets form a pinwheel-like structure while the moments on the central six-fold sites, which are frustrated due to the cancellation of interactions with nearest neighbours, do not order. The moments are ferromagnetically aligned along the *c* axis.

Immediately below the phase transition there is a significant increase in the DC and AC susceptibility (Mentink et al., 1996a) which is quenched by the application of a modest magnetic field (< 1 T). This is likely the signature of the ferromagnetically correlated chains on the non ordering sites. At low temperatures (< 2 K), however, this effect is eliminated, the resistivity passes through a maximum and c/T increases dramatically to over 500 $\mathrm{mJ/(mole\,K^2)}$ at 0.3 K (Mentink et al., 1996b). It appears that the frustration is being alleviated by the formation of heavy itinerant states without a moment in the presence of the localised moments which order at 21 K. This is similar to what occurs in $DyMn_2$ (Nuñez Regueiro and Lacroix, 1994) and CeSb (Ballou et al., 1991) and is the consequence of the combination of lattice frustration, a proximity to a magnetic-nonmagnetic transition and strong anisotropy.

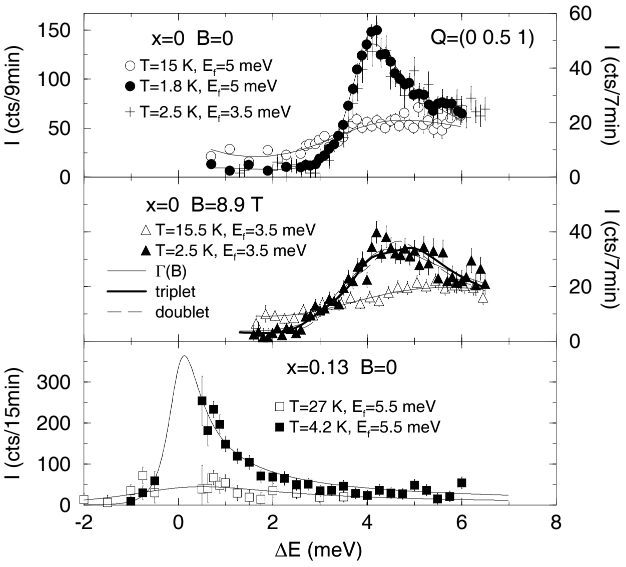

Figure 7. Constant-$\boldsymbol{Q}$ scans in CeNiSn for (upper panel) $B = 0$, (middle panel) $B = 8.9$ T and (lower panel) CeNi$_{0.87}$Cu$_{0.13}$Sn at $\boldsymbol{Q} = (0, 0.5, 1)$ showing the effect of increasing temperature and magnetic field on the inelastic response. From Schröder et al. (1996).

3 Semiconductors and Non-Fermi Liquids

3.1 CeNiSn

CeNiSn is interesting because it is the only Ce-based ‘Kondo insulator’ (for a review see Aeppli and Fisk, 1992) which can be readily fabricated in (large) single crystal form. Since the review of Aeppli and Broholm (1994), the material has received considerable attention from various groups throughout the world. The principal new results are:

(i) The discovery of a clean gapped signal at wavevectors of type $(0, 0.5, l)$ where l is an integer in addition to those equivalent to (0,0,1) (Kadowaki et al., 1994; Sato et al., 1995). Figure 7 shows the new peak, especially striking in its much more intense manifestation after the new Risø cold neutron guide tubes were installed (for a comparison between this spectrum and that taken before the installation of the new guides see Lebech (1993)). While the gap is larger (4 meV) at the former point than the latter (where it is 2 meV (Mason et al., 1992)), the property that $\chi''(\boldsymbol{Q}, \omega)$ is a strong function of $\boldsymbol{Q}$, while $\chi'(\boldsymbol{Q}, \omega = 0)$ is not, remains. Thus, the

puzzle of the 'shielded' RKKY interactions in Kondo insulators remains, although Varma (1995) has advanced arguments as to its resolution.
(ii) The discovery of long range magnetic order in CeNiSn samples doped by Cu substitution for Ni to achieve metallic heavy fermion behaviour. The magnetic ordering vector is close to the (0.5,l,0) vectors found to exhibit the higher gap frequency. Thus, in addition to producing a (dirty) metal, doping apparently eliminates the shielding phenomenon seen in the parent compound as well as the other celebrated single crystal Kondo insulator, FeSi.
(iii) The discovery that a magnetic field strongly affects the shape of the magnetic gap spectra (see Fig. 7). In particular, the gap appears less sharp, although one cannot judge whether this is due to field-induced splitting of some degeneracy or a true reduction in the lifetime of excitations at the gap energy. In spite of the considerable spectral change as well as the fact that the sample is rapidly approaching a metallic condition with increasing field, the shielding phenomenon mentioned in (i) and (ii) remains. In summary, the most important consequence of the new work on CeNiSn and its relatives is that there are dramatically different routes to metallic behaviour in heavy fermion systems, the first (doping) of which leads to substantial RKKY interactions while the second (external field) does not.

3.2 $\mathbf{Y_{1-x}U_xPd_3}$ and $\mathbf{UCu_{5-x}Pd_x}$

The properties of heavy fermion metals are a dramatic example of the success of Fermi liquid theory in the sense that the low temperature transport and thermodynamics, as well as the excitation spectra and quantum oscillations in a magnetic field are all in accord with predictions for a metal with a well defined Fermi surface (albeit with an extremely large effective mass due to electronic interactions). Similarly in Kondo insulators such as CeNiSn several distinct energy scales are directly manifested in the size of the gap and the properties of these materials are understandable in the framework of a band type picture even though more careful examination of the neutron data reveal important failings of band theory (Mason et al., 1992). There has been a great deal of interest recently in compounds, typically random alloys, which exhibit weak power law and logarithmic divergences in their low temperature properties at odds with the predictions of Fermi liquid theory, generically referred to as non-Fermi liquid (NFL) behaviour.

One such material is $\mathrm{Y_{1-x}U_xPd_3}$ with $x = 0.2$ (Seaman et al., 1991; Andraka and Tsvelik, 1991) which has a logarithmically diverging electronic specific heat below 20 K, a power law divergence of the susceptibility, and a resistivity which varies as $(1 - (T/T_o))^{1.13}$. This behaviour has been attributed to a two channel quadrupolar Kondo effect (Seaman et al., 1991), proximity to a novel zero temperature phase transition (Andraka and Tsvelik, 1991) or the suppression of the Kondo

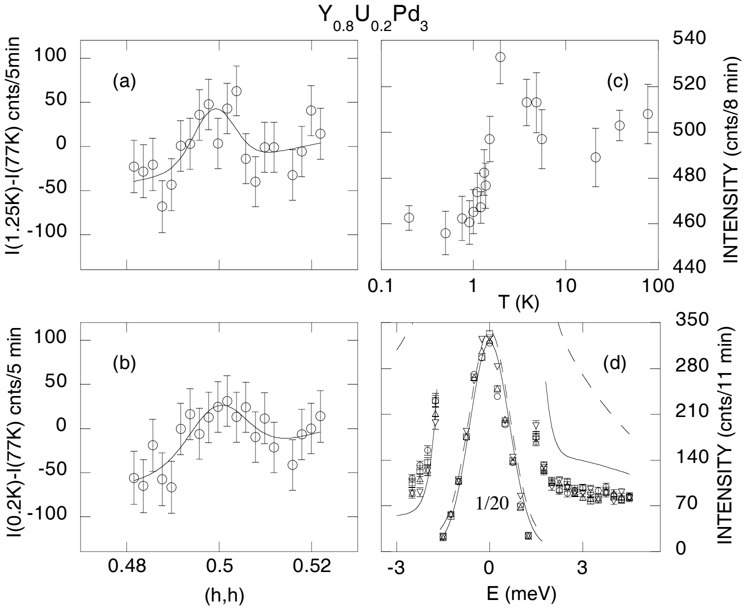

Figure 8. Magnetic correlations in $\mathrm{Y_{0.8}U_{0.2}Pd_3}$. (a) $\boldsymbol{Q}$ dependence of the energy integrated $S(\boldsymbol{Q},\omega)$ obtained by taking the difference in intensities at 1.25 K and 77 K. (b) The same difference between 0.2 K and 77 K. (c) Temperature dependence of the scattering at 0.5 meV for $\boldsymbol{Q} = (0.49, 0.49, 0)$. (d) Constant-$\boldsymbol{Q}$ scan at (0.5,0.5,0) at 70 K. From Dai et al. (1995)

temperature due to disorder and the associated proximity to a metal insulator transition (Dobrosavljević et al., 1992). Recent neutron scattering measurements by Dai et al. (1995) on polycrystalline samples with $x = 0.2$ and 0.45 have shed considerable light onto the ground state for this material. Figure 8 summarizes some of the results. Panels (a) and (b) show the weak peak in the energy integrated cross section which develops at low temperatures at the same antiferromagnetic wavevector at which long range order develops in the $x = 0.45$ compound (which had previously thought to be a spin glass). A temperature scan at 0.5 meV (panel (c)) shows a suppression of these fluctuations as the characteristic energy moves to lower energies below about 2 K. If the logarithmic increase in the resistivity in this material were due to the conventional Kondo effect then a quasielastic peak with a characteristic energy of $k_BT \sim 3.6$ meV would result in a constant-$\boldsymbol{Q}$ response shown as solid and dashed lines in panel (d), inconsistent with the data. Polarized beam measurements have shown that the dominant contribution to the magnetic scattering for both the $x = 0.2$ and $x = 0.45$ samples is a resolution limited re-

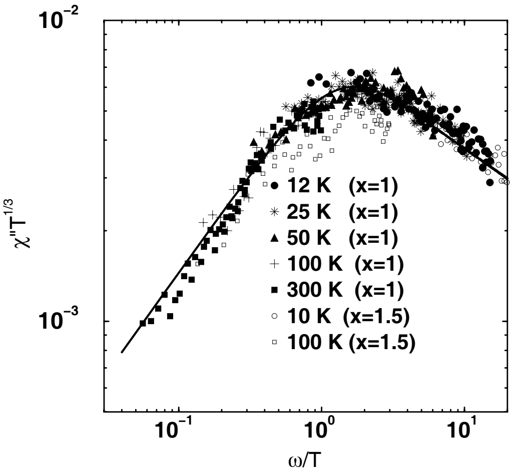

Figure 9. $UCu_{5-x}Pd_x$ exhibits scaling for both $x = 1$ and 1.5. The solid line corresponds to $\chi''(\omega, T)T^{1/3} \sim (T/\omega)^{1/3} \tanh(\omega/1.2T)$. From Aronson et al. (1995).

sponse centred on zero frequency. This indicates that in both cases the ground state for the U ions is the Γ_5 triplet. This magnetic ground state, suggested by the observation of weak critical scattering, rules out the quadrupolar two-channel mechanism for NFL behaviour in $Y_{1-x}U_xPd_3$.

Another instance of NFL behaviour occurs in $UCu_{5-x}Pd_x$. In this case there is randomness due to alloying as in $Y_{1-x}U_xPd_3$ however there is no site dilution of the U. The novel low temperature behaviour observed for $x = 1.5$ has been ascribed to the suppression of a spin glass transition to $T = 0$. Using time-of-flight powder measurements Aronson et al. (1995) have observed a magnetic excitation spectrum which, below a cross over of about 25 meV, is characterised by a scale which is determined by the temperature. At energies lower than T the dynamic susceptibility is proportional to ω/T, exactly cancelling the temperature factor and leading to a temperature independent cross section, $S(\omega)$ similar to what has been seen in lightly doped cuprates (Hayden et al., 1991). This scaling behaviour, which is the same for $x = 1$ and 1.5, is explicitly shown in Fig. 9. Surprisingly for a dense Kondo lattice, there is no observable Q dependence other than the overall form factor dependence. This could suggest a single ion origin for the observed scaling although it may also be due to the directional averaging which occurs in any powder measurement. As in $Y_{1-x}U_xPd_3$, the quasielastic response indicates a magnetic ground state which has an instability driven towards $T = 0$.

The novel effects seen in these materials are not limited to alloys with compositional disorder. Similar effects are seen in URh_2Ge_2 (Süllow et al., 1996) although substitutional disorder between Rh and Ge likely plays a role. In that case there is clearly a competition between spin glass and antiferromagnetic order which may drive the low temperature properties.

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