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1996 J. Phys.: Condens. Matter 8 9855

(http://iopscience.iop.org/0953-8984/8/48/013)

J. Phys.: Condens. Matter **8** (1996) 9855–9870. Printed in the UK

NMR and μSR studies of non-Fermi-liquid behaviour in disordered heavy-fermion systems

D E MacLaughlin†, O O Bernal‡ and H G Lukefahr§

† Department of Physics, University of California, Riverside, CA 92521, USA
‡ Department of Physics, California State University, Los Angeles, CA 90032-8206, USA
§ Whittier College, Whittier, CA 90608, USA

Received 24 July 1996, in final form 11 September 1996

Abstract. Evidence has emerged from nuclear magnetic resonance (NMR) and muon spin-rotation (μSR) experiments that non-Fermi-liquid (NFL) behaviour in heavy-fermion alloys can in some cases be due to an inhomogeneous distribution of Kondo temperatures T_K arising from disorder in the random alloy. Such 'Kondo disorder' implies a broad distribution of the heavy-fermion spin polarization, which is reflected in the widths of NMR and μSR lines. A simple model for the shape and width of the distribution of T_K accounts for the temperature and field dependence of the bulk susceptibility in the NFL alloys $UCu_{5-x}Pd_x$, $x = 1.0$ and 1.5, and then agrees with NMR values of the width $(\delta\chi)_{\rm rms}$ of the susceptibility distribution with no further adjustable parameters. Comparison of NMR and μSR estimates of $(\delta\chi)_{\rm rms}$ indicates that the susceptibility is disordered on an atomic length scale. In contrast, μSR lines in $CeCu_{5.9}Au_{0.1}$ are too narrow for Kondo disorder to account for NFL properties unless, as seems unlikely, the correlation length is long in this alloy. Similarly, small low-temperature ^{89}Y NMR linewidths make it unlikely that Kondo disorder is the origin of NFL behaviour in $Y_{0.8}U_{0.2}Pd_3$.

1. Introduction

Nozières [1] was the first to suggest that low-lying excitations in Kondo alloys can be described by Landau's Fermi-liquid theory, in which there is a one-to-one correspondence between the excitations of the correlated-electron system and those of a free-electron gas. Signatures of this Fermi-liquid behaviour are usually taken to be the well-known low-temperature thermal and transport properties of a Fermi liquid: the susceptibility $\chi(T)$ and Sommerfeld (T-linear) coefficient $\gamma(T) = C(T)/T$ of the specific heat both become constant as $T \to 0$, and the resistivity $\rho(T)$ varies asymptotically as T^2. Fermi-liquid behaviour has also become the canonical description of concentrated Kondo or heavy-fermion systems, principally alloys and intermetallic compounds of lanthanide (Ce, Yb) and actinide (U) ions.

A large and growing number of heavy-fermion alloys [2, 3] are not described by this Fermi-liquid picture. A review of the current situation and a list of the so-called non-Fermi-liquid (NFL) heavy-fermion alloys known of to date is given by Maple elsewhere in this Special Issue [4]. Thermal and transport properties of NFL materials as $T \to 0$ seem to fall into a limited number of classes; most (but not all) exhibit a logarithmic divergence of $\gamma(T)$ and a linear departure of $\rho(T)$ from its value at $T = 0$. The susceptibility has been reported to vary either as $1 - b(T/T_K)^{1/2}$ or as $-\ln(bT/T_K)$, where T_K is the Kondo temperature and b is a dimensionless factor of order unity. Nearly all NFL heavy-fermion

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materials are disordered alloys, and all are found in the neighbourhood of a transition to magnetic order in a temperature–composition phase diagram. Both Ce- and U-based alloys exhibit NFL behaviour, and it is found both with and without a disordered f sublattice.

A number of mechanisms for NFL behaviour have been proposed, of which two, discussed at length in other contributions to this Special Issue, have received the most attention to date. These are (a) the multichannel Kondo effect, first associated with a two-channel quadrupolar Kondo mechanism by Cox [5] and since extended to other mechanisms for two-channel Kondo screening [6], and (b) magnetic instability due to a quantum critical point at zero temperature [7, 8], the critical behaviour of which generates NFL properties. This paper concentrates on a third possible mechanism: an inhomogeneous distribution of Kondo temperatures due to structural disorder in the alloy, referred to hereafter as 'Kondo disorder'.

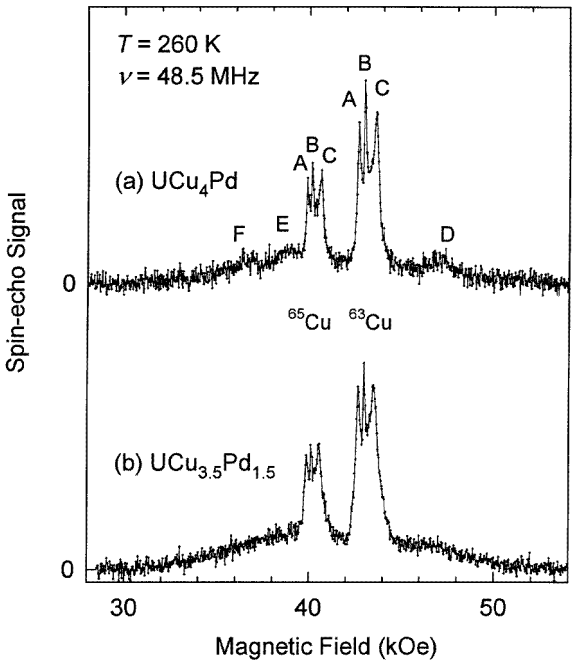

Figure 1. Copper NMR spectra for $UCu_{5-x}Pd_x$, $x = 1.0$ (a) and 1.5 (b), $T = 260$ K. The prominent peaks are the ^{63}Cu and ^{65}Cu central transitions. A and C: powder-pattern singularities. B: lines from a **Cu**Pd impurity phase. D–F: quadrupole-split satellites, further broadened in $UCu_{3.5}Pd_{1.5}$. Data from reference [19].

Kondo disorder has been treated theoretically in a number of contexts [9–13]. Its application to heavy-fermion NFL alloys was motivated by the observation of broad nuclear magnetic resonance (NMR) [14] and muon spin-rotation (μSR) [15] lines in $UCu_{5-x}Pd_x$, $x = 1.0$ and 1.5. These large linewidths are unambiguously due to static disorder, since the separately measured dynamic (lifetime) broadening is much less than the observed widths, and indicate the presence of an inhomogeneous distribution of the static spin polarization associated with the U ions. Zero-field μSR experiments on a number of NFL alloys [15–17] show no static magnetic moment greater than $\sim 0.01\,\mu_B$/f ion, so the spin polarization is paramagnetic in origin and not due to static magnetism associated

with antiferromagnetic or spin-glass freezing of the U-ion spins. The inhomogeneous spin polarization is therefore proportional to the applied field, and can be characterized by a corresponding inhomogeneous susceptibility.

2. Copper NMR in $UC_{5-x}Pd_x$: Kondo disorder?

We begin by describing features of copper NMR spectra of the NFL alloys $UCu_{5-x}Pd_x$, $x = 1.0$ and 1.5, in particular the additional inhomogeneous broadening which suggests Kondo disorder.

If the resonant nucleus is not at a site of tetrahedral or higher point symmetry, then the anisotropy of the crystalline environment leads to NMR frequencies which depend on the orientation of the crystal relative to the applied magnetic field [18]. If in addition the nucleus possesses an electric quadrupole moment, the electric field gradient at the nuclear site splits the NMR spectrum into more than one line, the frequencies of which are also anisotropic. Metallic samples for NMR experiments are usually powdered to permit penetration of the radio-frequency magnetic fields required to carry out the NMR study. The powder grains are generally randomly oriented, so the anisotropy and quadrupole splittings give rise to a characteristic 'powder-pattern' lineshape [18], consisting of a central transition and quadrupole satellites.

Figure 1 gives representative copper NMR spectra from $UCu_{5-x}Pd_x$, $x = 1.0$ and 1.5, at $T = 260$ K. The two prominent lines are central transitions from the two naturally abundant ^{63}Cu and ^{65}Cu isotopes of copper. A powder-pattern background of quadrupole satellites is also visible. Our $UCu_{5-x}Pd_x$ samples were slightly contaminated with a few per cent of a **Cu**Pd second phase, which gave rise to extra Cu NMR lines (one for each isotope). All of these features can be seen in figure 1 [19]. The broadening of interest is in addition to the powder-pattern broadening and is most visible at low temperatures; an example at 10.5 K is given in figure 2. We emphasize that this additional broadening could not be present in a perfect crystal, for which all Cu sites would be equivalent, and must therefore be due to disorder.

The additional NMR broadening suggests a broad distribution of Kondo temperatures T_K, as can be seen from the following crude argument. For a given value of T_K the corresponding susceptibility $\chi(T; T_K)$ is roughly Curie–Weiss-like:

$$\chi(T; T_K) = \mathcal{C}/(T + \alpha T_K) \tag{1}$$

where $\mathcal{C}$ is the free-spin Curie constant and α is a factor of order unity. Then for a temperature-independent distribution function $P(T_K)$ which describes the disorder in T_K the corresponding distribution function $P(\chi)$ for the susceptibility will be broader at low temperatures, where the temperature T in equation (1) is small and unimportant, than at high temperatures, where it is T_K which becomes unimportant. This behaviour agrees with that shown in figures 1 and 2, where the line broadening increases as the temperature is reduced, and motivates consideration of whether Kondo disorder could give rise to the NFL behaviour of $UCu_{5-x}Pd_x$.

3. Kondo disorder and NFL behaviour

Since NFL behaviour involves low-lying excitations, any mechanism giving rise to it must affect the smallest energies in the system. We therefore assume that the distribution function $P(T_K)$ is broad enough that $P(T_K{=}0) \neq 0$, i.e., that there are a finite number of Kondo spins with $T_K < T$ at any finite temperature T. We can think of the

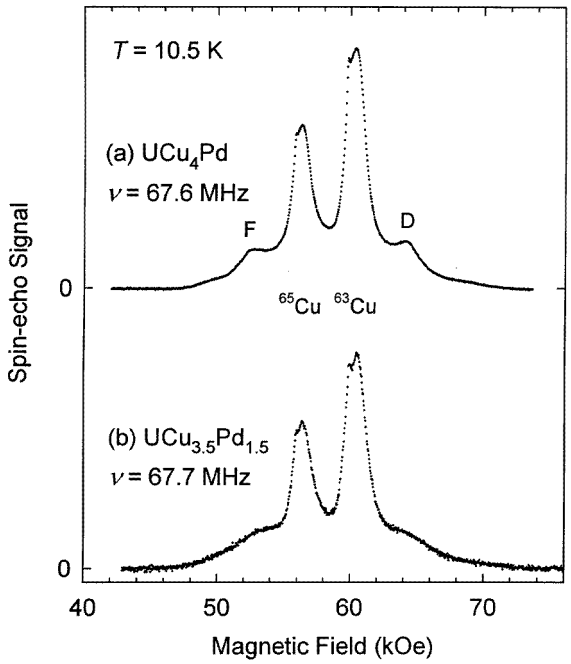

Figure 2. Copper NMR spectra for $UCu_{5-x}Pd_x$, $x = 1.0$ (a) and 1.5 (b), $T = 10.5$ K. Features are labelled as in figure 1. The powder-pattern structure exhibits additional broadening at low temperatures. Data from reference [19].

Kondo singlet state as broken up for these spins; they become 'free', dominate the low-temperature thermodynamics and transport properties, and give rise to NFL behaviour precisely because they are not in the low-temperature Kondo-compensated Fermi-liquid regime ($T \ll T_K$). In the Kondo-disorder model the magnetization $M(H, T)$ of the low-T_K free spins approximately follows a Brillouin function, and saturates in a sufficiently strong magnetic field $H \gg k_B T/\mu_B$. This leads to a field dependence of the sample-averaged or bulk magnetic susceptibility $\langle\chi(H, T)\rangle = \langle M(H, T)\rangle/H$.

We take this picture further by constructing a simple model of Kondo disorder from a microscopic point of view [14]. The Kondo temperature is given roughly by

$$T_K = E_F \exp(-1/\lambda)$$

where E_F is the Fermi energy and $\lambda = \rho\mathcal{J}$ is the Kondo coupling constant; here ρ is the density of conduction-band states at the Fermi energy and $\mathcal{J}$ is the conduction-electron–Kondo-ion exchange constant. Even modest disorder in either ρ or $\mathcal{J}$ produces considerable disorder in T_K, thanks to the amplifying effect of the exponential dependence for small λ. We assume a relatively narrow Gaussian distribution function $P(\lambda)$ for the distribution of λ; as shown below we find widths $w = (\delta\lambda)_{\rm rms}$ of the order of 20% of the mean $\langle\lambda\rangle$ in $UCu_{5-x}Pd_x$ for $x = 1.0$ and 1.5.

For these alloys the resulting distribution functions $P(T_K) = |\mathrm{d}\lambda/\mathrm{d}T_K|P(\lambda)$, shown in figure 3, are quite broad, and $P(T_K = 0) \neq 0$ as required to obtain asymptotic NFL behaviour. Although a distribution of U-ion crystal-field splittings or exchange interactions could also result in a corresponding distribution of Curie–Weiss temperatures as in equation (1), it would be hard to smear them out this much with structural disorder; the

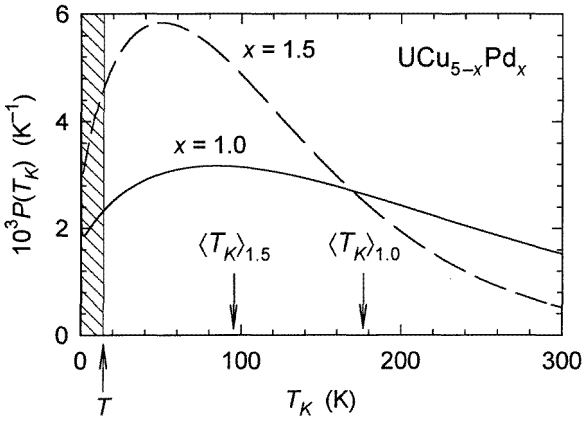

Figure 3. Distribution functions $P(T_K)$ of Kondo temperatures T_K in the Kondo-disorder model, calculated using parameters from fits to the field- and temperature-dependent susceptibility of UCu_4Pd and $UCu_{3.5}Pd_{1.5}$ as described in the text. The average Kondo temperatures $\langle T_K\rangle$ are shown for each alloy. For a given (low) temperature T the shaded area indicates the values of T_K for which the Kondo spins are uncompensated, i.e., 'free'.

exponential dependence of T_K on λ provides the needed spread in $P(T_K)$. Furthermore, there is no evidence for either crystal-field splittings or exchange interactions with characteristic energies in the required range (100–200 K; see section 5). In particular, exchange interactions would result in the onset of static magnetism at temperatures of this order contrary to experiment [15].

For comparison with experiment we wish to calculate the average and rms width of the susceptibility distribution. The general nth moment is given by

$$\langle[\chi(H,T)]^n\rangle = \int \mathrm{d}^3r\,[\chi(\boldsymbol{r})]^n = \int_0^\infty \mathrm{d}T_K\; P(T_K)[\chi(H,T;T_K)]^n \tag{2}$$

and we make a crude representation of the single-ion Kondo physics by writing the susceptibility $\chi(H,T;T_K) = M(H,T;T_K)/H$ as

$$\chi(H,T;T_K) = g\mu_B J B_J(x)/H. \tag{3}$$

Here $B_J(x)$ is the Brillouin function for free spins of angular momentum J, and

$$x = \frac{g\mu_B JH}{k_B(T+\alpha T_K)}.$$

This preserves the approximate Curie–Weiss form of the Kondo susceptibility, and also allows low-temperature saturation of spins with small T_K.

Our analysis of Kondo disorder proceeds as follows. We first obtain the parameters of $P(T_K)$ (the mean and width of the distribution of $P(\lambda)$) by fitting the bulk average susceptibility (first moment) $\langle\chi(H,T)\rangle$ to the experimental data [14]. The field dependence of χ gives the needed sensitivity to the width of $P(\lambda)$. Once we have defined $P(T_K)$ we can calculate the rms width $(\delta\chi)_{\mathrm{rms}} = (\langle\chi^2\rangle - \langle\chi\rangle^2)^{1/2}$ with no further adjustable parameters. This theoretical width can then be compared with estimates of $(\delta\chi)_{\mathrm{rms}}$ obtained from NMR and μSR linewidths as described in the next section.

4. The spin-probe linewidth and the inhomogeneous susceptibility

In a solid with paramagnetic spins, such as a heavy-fermion alloy, a *spin probe* (nuclear or muon spin) precesses in the sum of the applied static field and any extra field due to the static spin polarization. The relative shift (the Knight shift in metals) $K_i = \Delta\nu_i/\nu_{\rm ref}$ of the precession frequency ν_i of the ith probe spin is given by [18]

$$K_i = \sum_j a_{ij}\chi_j \tag{4}$$

where χ_j is the local susceptibility associated with the jth paramagnetic ion, and a_{ij} is the coupling constant which describes the dipolar and/or transferred hyperfine interaction between probe spin i and paramagnetic ion j. Any distribution of the χ_j produces a corresponding distribution of the K_i, and the NMR line will be broadened. It is shown in appendix A (see also reference [15]) that the relative spread $(\delta\chi)_{\rm rms}/\langle\chi\rangle$ in the susceptibility is related to the relative linewidth $\kappa = (\delta K)_{\rm rms}$ by

$$(\delta\chi)_{\rm rms}/\chi = \kappa/(a^*\chi) \tag{5}$$

(see equation (A2)) where $\chi = \langle\chi\rangle$ and a^* is an effective coupling constant, which can be obtained [15] from the calculated dipolar coupling and a separate measurement of the average NMR shift $\langle K\rangle$.

It is shown in appendix A that when the χ_j are disordered a^* depends on the correlation length ξ associated with this disorder: $a^* = a^*(\xi)$ [15]. There are two extreme cases.

- *Long*-range correlation (LRC) ($\xi \gg$ lattice constant): each probe spin sees an essentially uniform neighbour Kondo-ion polarization which is, however, different for different probe-spin sites. In this case $a^* = a^*_{\rm LRC} = |\sum_j a_{ij}|$ (equation (A3)).
- *Short*-range correlation (SRC) ($\xi \lesssim$ lattice constant): each probe spin sees an essentially uncorrelated distribution of neighbouring Kondo-ion polarizations. In this case $a^* = a^*_{\rm SRC} = (\sum_j a_{ij}^2)^{1/2}$ (equation (A4)).

In other words, cross terms in the sum are important in the LRC limit but are absent in the SRC limit. Clearly $a^*_{\rm SRC}$ may be quite different from $a^*_{\rm LRC}$.

One cannot obtain ξ from a single spin-probe experiment. But it may be possible to estimate the correlation range crudely (i.e., to determine which limit, SRC or LRC, is more appropriate) by comparing data from different spin probes, e.g., from NMR and μSR. The procedure is to look for agreement between values of $\kappa/(a^*\chi)$ as obtained from different spin probes but in the same correlation limit. Because $a^*_{\rm LRC} \neq a^*_{\rm SRC}$ for a given spin probe, and because the a^* are also different for different spin probes, one expects agreement between the spin-probe values of $\kappa/(a^*\chi)$ only for the appropriate correlation limit.

In the above we have assumed that the Kondo-ion polarizations are disordered but the coupling constants are not, i.e., a_{ij} has the same value for all crystallographically equivalent sites i and j. The NMR linewidth for the case where both χ and a_{ij} are disordered is discussed in appendix B. In the case where χ is uniform but a_{ij} is distributed we have $\langle K\rangle \propto \langle\chi\rangle$ and $\langle K^2\rangle \propto \langle\chi\rangle^2$, so $\kappa \propto \langle\chi\rangle$; $\kappa/(a^*\chi) =$ constant. We have seen above that $\kappa/(a^*\chi) = (\delta\chi)_{\rm rms}/\chi$ if the broadening is due to a distribution of χ. Thus a constant $\kappa/(a^*\chi)$ is either a signature of disorder in a_{ij} or, alternatively, an indication that $(\delta\chi)_{\rm rms}$ is proportional to χ. No microscopic mechanism for such a proportionality suggests itself, but it does occur for macroscopic sources of inhomogeneity (non-uniform dipolar or demagnetizing fields). These can be estimated and turn out not to be important. In a plot of $\kappa/(a^*\chi)$ versus χ, with temperature an implicit parameter, we will find $\kappa/(a^*\chi) =$ constant

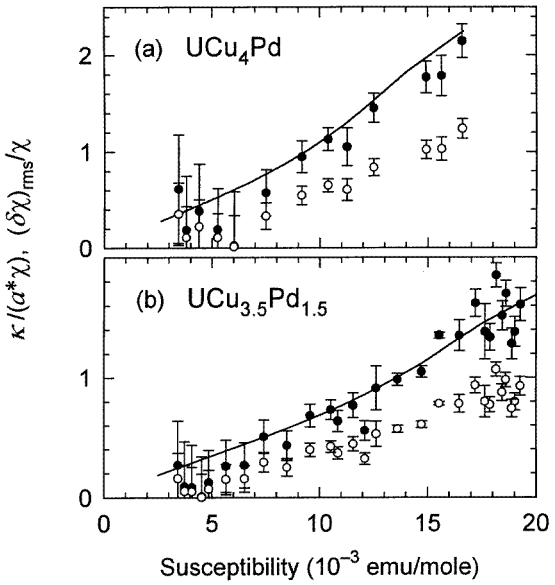

Figure 4. The dependence of the relative copper NMR linewidth $\kappa/(a^*\chi)$ on bulk susceptibility, with temperature an implicit parameter, in (a) UCu_4Pd and (b) $UCu_{3.5}Pd_{1.5}$. Filled circles: a^* evaluated in the limit of short correlation length. Open circles: a^* evaluated in the limit of long correlation length. Curves: relative rms widths $(\delta\chi)_{\rm rms}/\chi$ of susceptibility distributions from a Kondo-disorder model using $P(T_K)$ shown in figure 3. Data from reference [14].

if the static broadening is due to a distribution of a_{ij}. Observation of a χ-dependent $\kappa/(a^*\chi)$, however, implies new physics.

Figure 4 gives the dependence of $\kappa/(a^*\chi)$ on χ for $UCu_{5-x}Pd_x$, $x = 1.0$ and 1.5, evaluated using Cu NMR data and calculated values [14, 15] of $a^*_{\rm SRC}$ and $a^*_{\rm LRC}$. In both correlation length limits $\kappa/(a^*\chi)$ is found to be strongly χ dependent—large for large χ (low temperatures) and vanishing for small χ; the latter rules out a significant distribution of a_{ij}. We conclude that $\kappa/(a^*\chi)$ is a measure of $(\delta\chi)_{\rm rms}/\chi$ in these alloys. It is a dimensionless number which becomes $\gtrsim 1$ at low temperatures (large χ). This means that the distributions of susceptibility are very broad, with rms widths of the order of the average.

Thus disorder in χ is important (and comparable in size) in both $UCu_{3.5}Pd_{1.5}$ and UCu_4Pd. It has been suggested, however, that UCu_4Pd is an ordered stoichiometric compound [20]. The broad NMR lines in this alloy can only arise from some kind of inhomogeneity, so there must be Cu–Pd site interchange or another source of structural disorder. We know of no evidence against such disorder from x-ray or neutron Bragg scattering, but more detailed studies are needed. We also note that the magnetic susceptibility (figure 5) was measured before and after grinding the sample into a powder and found to be unchanged; any modification of $P(T_K)$ due to disorder introduced by the grinding was therefore negligible.

5. Analysis of Kondo disorder in $UCu_{5-x}Pd_x$

We now proceed to test the Kondo-disorder model using the procedure of section 3. First, the experimental field and temperature dependence of the bulk susceptibility $\chi(H, T)$ are fitted

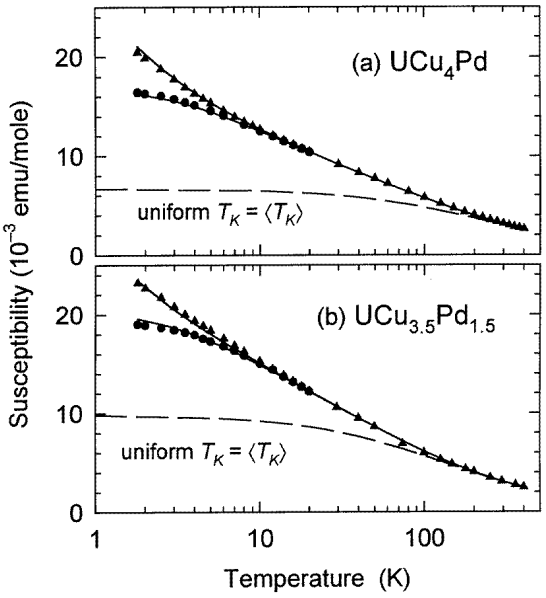

Figure 5. The temperature dependence of the bulk magnetic susceptibility of (a) UCu_4Pd and (b) $UCu_{3.5}Pd_{1.5}$. Triangles: $H = 5$ kOe. Circles: $H = 50$ kOe. Solid curves: fits to the Kondo-disorder model (see the text). Dashed curves: no Kondo disorder (uniform $T_K = \langle T_K \rangle$). From reference [14].

to the model function, defined by equation (2) (with $n = 1$) and equation (3). We note again that the field dependence in this model arises from the saturation of the uncompensated spins ($T > T_K$) for high enough field. The results of this procedure for UCu_4Pd and $UCu_{3.5}Pd_{1.5}$ are shown in figure 5, where it can be seen that the fit is quite good. The resulting distribution functions $P(T_K)$ have been given in figure 3. For the parameters of $P(\lambda)$ we find $\langle\lambda\rangle \approx 0.2$, corresponding to an average Kondo temperature $\langle T_K \rangle \approx 100$–$200$ K, and $w \approx 0.04$. The behaviour of $\chi(T)$ is very different from that for the same mean Kondo temperature but $w = 0$, given by the dashed lines in figure 5. The good fits are consistent with Kondo disorder but not particularly strong evidence for it, since the low-temperature increase and field dependence of $\chi(T)$ could be due to isolated impurity spins, spins in small quantities of a second metallurgical phase, etc.

We next calculate $(\delta\chi)_{\rm rms}/\chi$, using equation (2) with $n = 1$ and 2 and parameters from the fit to the susceptibility. The results are given by the curves in figure 4, which can be compared with $\kappa/(a^*\chi)$ from the NMR linewidths. There is a factor-of-two agreement with LRC-limit data analysis ($a^* = a^*_{\rm LRC}$) for both $x = 1.0$ and 1.5, and even better agreement in the SRC limit ($a^* = a^*_{\rm SRC}$). This is strong evidence that Kondo disorder contributes to NFL behaviour in $UCu_{3.5}Pd_{1.5}$ and UCu_4Pd. It can also be taken as evidence that the SRC limit is applicable, although this should be considered very tentative as it relies on comparison with theory rather than being a direct consequence of experimental results.

We can also compare the Kondo-disorder prediction with experimental data for other properties. As an example we have calculated the Kondo-disorder-model specific heat [14] using the simple 'resonant-level' theory of Schotte and Schotte [21]. Here $C = C(H, T; \Delta)$, where $\Delta = \beta T_K$ is the width of the resonant level and is a distributed quantity in the Kondo-disorder picture. The coefficient of proportionality β was taken as an additional fit

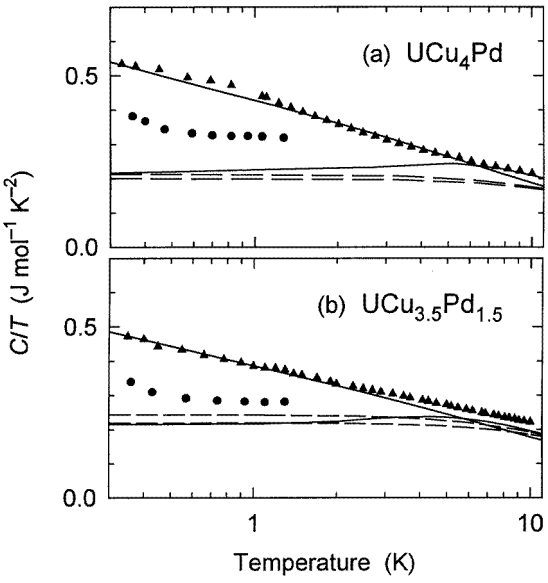

Figure 6. The temperature dependence of the specific heat coefficient $C(T)/T$ of (a) UCu_4Pd and (b) $UCu_{3.5}Pd_{1.5}$. Triangles, upper curves: $H = 0$. Circles, lower curves: $H = 140$ kOe. Solid curves: the Kondo-disorder model (see the text). Dashed curves: no Kondo disorder (uniform $T_K = \langle T_K \rangle$). From reference [14].

parameter, and was found to be $\approx 1/2$ for the best fit to specific heat data for $H = 0$. The fits are shown in figure 6. Rough agreement is obtained without further adjustment of other parameters. The field dependence of the specific heats, also shown in figure 6, are not in good agreement with the calculation. This may be a failure of the resonant-level model, and comparison with a better theory such as the Bethe *ansatz* solution should be carried out.

A preliminary comparison of the neutron inelastic scattering data of Aronson *et al* [22] with the Kondo-disorder model is reported by Miranda *et al* in this Special Issue [13], and will not be discussed in detail here. Although Aronson *et al* interpreted their data in terms of a scaling picture which is not strictly obeyed by the Kondo-disorder model, the discrepancy turns out to be small enough so that the neutron scattering results do not rule out Kondo disorder.

6. μSR in $UCu_{5-x}Pd_x$

We return to the question of the correlation length for the disordered susceptibility in $UCu_{5-x}Pd_x$. It so happens that μSR in these alloys is particularly helpful due to the following special circumstance. The point symmetry of both known muon stopping sites in the isostructural end compound UCu_5 [23] is tetrahedral ($\bar{4}3m$), and for this symmetry the contribution to a_{LRC} from dipolar fields vanishes if the susceptibility in the neighbourhood of the site is uniform. It is also known that the muon/U-ion coupling is predominantly dipolar in UCu_5 (and by extension $UCu_{5-x}Pd_x$), as the transferred hyperfine interaction is weak in comparison. Then one expects a broad μSR line from susceptibility inhomogeneity only if the susceptibility is locally disordered, i.e., in the SRC limit. Put another way, a

given observed μSR linewidth requires a much broader susceptibility distribution in the LRC limit than in the SRC limit.

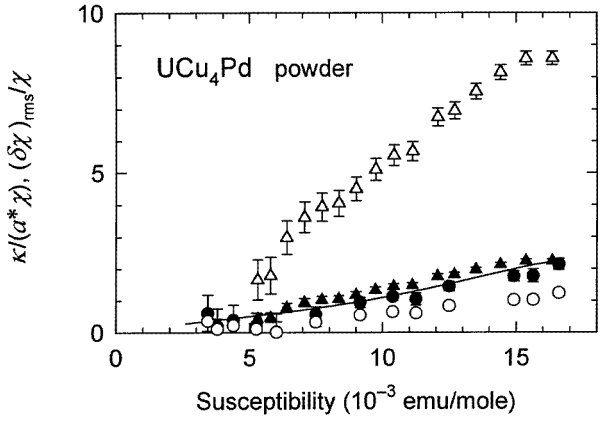

Figure 7. The dependence of relative linewidths $\kappa/(a^*\chi)$ on the bulk susceptibility in UCu_4Pd. Circles: NMR linewidths (data from reference [14]). Triangles: μSR linewidths (data from reference [15]). Filled symbols: a^* evaluated in the limit of short correlation length. Open symbols: a^* evaluated in the limit of long correlation length. Curve: the relative rms width $(\delta\chi)_{\rm rms}/\chi$ of the susceptibility distribution from the Kondo-disorder model using $P(T_K)$ shown in figure 3.

This can be seen in figure 7, which gives the dependence of $\kappa/(a^*\chi)$ in UCu_4Pd from NMR and μSR data with a^* evaluated in both the LRC and SRC limits. It can also be seen that the NMR and μSR values of $\kappa/(a^*\chi)$ are in good agreement with each other with a^* evaluated in the SRC limit (filled points), but that the agreement is very poor in the LRC limit (open points). This is excellent evidence that the SRC limit is appropriate to this alloy; a conclusion which, we stress, emerges solely from analysis of the linewidths and does not rely on the validity of the Kondo-disorder model. But then very good agreement with $(\delta\chi)_{\rm rms}/\chi$ from the Kondo-disorder model (the curve in figure 7) is found independently. This agreement constitutes the most persuasive evidence for the applicability of the Kondo-disorder picture in this case.

7. μSR in $CeCu_{5.9}Au_{0.1}$

There is a considerable body of evidence [24] that NFL behaviour in $CeCu_{5.9}Au_{0.1}$ is associated with a quantum critical point at zero temperature. It is nevertheless important to determine whether Kondo disorder plays a role, and to this end we have carried out μSR experiments on a single crystal of this alloy.

Unfortunately a comparison between μSR and NMR data as described above for $UCu_{5-x}Pd_x$ cannot be made as the NMR data are not available. Copper NMR in $CeCu_{5.9}Au_{0.1}$ is possible (Ce and Au isotopes have either vanishing or prohibitively weak nuclear moments) but will be difficult to interpret because the crystal structure is complex; there are five crystallographically inequivalent copper cites in the unit cell. One then expects at least thirty separate NMR spectral lines from the two copper isotopes, corresponding to different Knight shifts and quadrupole splittings. Copper NMR experiments in field-aligned powders of $CeCu_{5.9}Au_{0.1}$ are currently in progress.

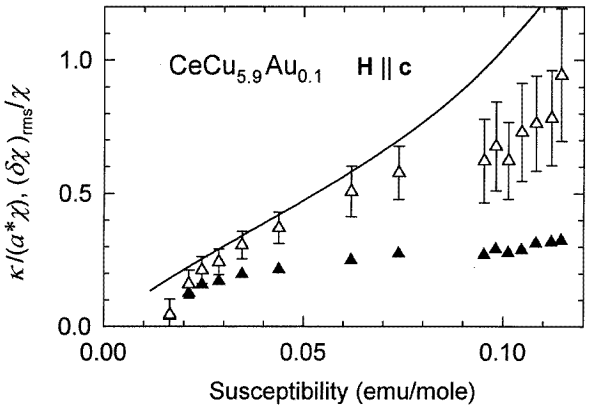

Figure 8. The dependence of the relative linewidths $\kappa/(a^*\chi)$ on the bulk susceptibility in $CeCu_{5.9}Au_{0.1}$. Filled symbols: a^* evaluated in the limit of short correlation length. Open symbols: a^* evaluated in the limit of long correlation length. Curve: the relative rms width $(\delta\chi)_{rms}/\chi$ of the susceptibility distribution from the Kondo-disorder model. Data from reference [15].

Figure 8 gives the dependence of $\kappa/(a^*\chi)$ on χ in $CeCu_{5.9}Au_{0.1}$, obtained from μSR linewidths in both the SRC and LRC limits. (The reduction of $\kappa/(a^*\chi)$ for small χ, i.e., at high temperature, is probably due to motional narrowing of the line by thermally activated muon diffusion, and is therefore an artifact of the μSR technique.) It can be seen that as in $UCu_{5-x}Pd_x$ the results depend markedly on the assumed correlation length limit. In the LRC limit $\kappa/(a^*\chi)$ becomes of order unity for large χ, as in $UCu_{5-x}Pd_x$, but in the SRC limit $\kappa/(a^*\chi)$ is considerably smaller and not as temperature dependent apart from the small-χ reduction.

This suggests that if the SRC limit is appropriate, then the μSR linewidth may be dominated by a distribution of coupling constants a_{ij} rather than by a distribution of susceptibilities. In other words Kondo disorder is at most present at a low level in $CeCu_{5.9}Au_{0.1}$, and is not responsible for its NFL properties. This conclusion is reinforced by comparison with $(\delta\chi)_{rms}/\chi$ predicted from the Kondo-disorder model (the curve in figure 8), using the temperature and field dependence of the bulk susceptibility as described above. If on the other hand the LRC limit is appropriate, then $\kappa/(a^*\chi)$ is comparable to the Kondo-disorder-model prediction.

Because NMR data are not available we cannot distinguish between LRC and SRC limits directly, and we are forced to consider less direct evidence. It has been noted [25] that many properties of $CeCu_{6-x}Au_x$ can be understood in a two-component model for the magnetic behaviour of the Ce ions, where the two components differ in whether or not an Au atom is a near neighbour to a Ce site. Such a picture implies short-range correlation of any disorder in χ. Then Kondo disorder is insufficient to account for the NFL behaviour, a conclusion which is in line with the magnetic instability scenario.

8. NMR in $Y_{1-x}U_xPd_3$

In both $UCu_{5-x}Pd_x$ and $CeCu_{5.9}Au_{0.1}$ disorder is associated with the non-f ligand ions; the f sublattice remains periodic. In the NFL heavy-fermion alloy series $Y_{1-x}U_xPd_3$, however, the f sublattice is diluted. These NFL alloys exhibit one signature of Kondo disorder,

namely, saturation of the magnetization with field at low temperatures. As noted above this is not a sufficient condition for the applicability of the Kondo-disorder model, since impurity spins or second phases could also yield a saturating magnetization. We therefore examine the behaviour of the ^{89}Y NMR linewidth in $Y_{1-x}U_xPd_3$ [26] for signs of Kondo disorder.

For $T \gtrsim T_K$ the ^{89}Y line is Gaussian, with a linewidth proportional to χ. This proportionality is a sign of disorder in coupling constants a_{ij}, not in χ, as described above, and is expected: even if the U-ion susceptibility was uniform, the random occupancy of f-sublattice sites by U and Y ions can be thought of as turning off the interaction between a spin probe and those neighbouring sites which are occupied by Y ions. As shown in appendix B, this builds in a contribution to the linewidth proportional to χ for dilute f alloys.

For $T \lesssim T_K$, however, the lineshape changes, and becomes nearly Lorentzian at low temperatures. We have chosen to characterize this change in lineshape by measuring the width of the field-swept NMR spectrum at two points relative to the maximum. In addition to the usual half-width at half-maximum $(\Delta H)_{1/2}$, we also measure the half-width at *tenth*-maximum $(\Delta H)_{1/10}$. Since Gaussian and Lorentzian lineshapes differ in that more of the spectral weight is in the 'shoulders' of the line for the Lorentzian, the ratio $(\Delta H)_{1/10}/(\Delta H)_{1/2}$ is larger for a Lorentzian line than for a Gaussian line.

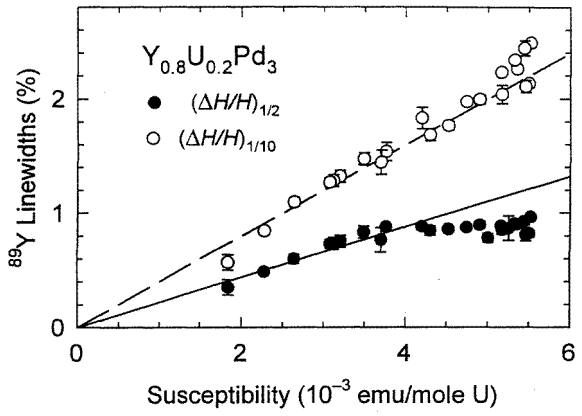

Figure 9. The dependence of the ^{89}Y NMR linewidths on the bulk susceptibility in $Y_{0.8}U_{0.2}Pd_3$. Filled symbols: full width at half-maximum $(\Delta H/H)_{1/2}$. Open symbols: full width at tenth-maximum $(\Delta H/H)_{1/10}$. Data from reference [26].

The dependence of the relative linewidths $(\Delta H/H)_{1/2}$ and $(\Delta H/H)_{1/10}$ on the bulk susceptibility in $Y_{0.8}U_{0.2}Pd_3$ is given in figure 9. It can be seen that $(\Delta H/H)_{1/10}$ remains proportional to χ, whereas $(\Delta H/H)_{1/2}$ falls *below* the extrapolated high-temperature proportionality to χ. The mechanism for this behaviour is unclear [26]; for the present purposes it is enough to note that the dependence of the NMR linewidth on χ is linear or slower.

In appendix B we show that assuming the SRC limit for the distribution of the a_{ij} as well as of χ, and assuming furthermore that these two distributions are not correlated with each other, leads to contributions to the spin-probe linewidth which add in quadrature (equation (B2)). Under these assumptions, therefore, the contribution from Kondo disorder, expected to be most important at low temperatures, can only increase κ over its high-

temperature value determined by the distribution of a_{ij}.

If these assumptions are valid, therefore, the observed *decrease* of $(\Delta H/H)_{1/2}$ at low temperatures is evidence that Kondo disorder is not the source of NFL behaviour of $Y_{0.8}U_{0.2}Pd_3$. In this case the field dependence of χ must be ascribed to an impurity contribution, which should be subtracted [27] to obtain the intrinsic susceptibility. Little is known concerning correlations between χ and a_{ij}, however, so Kondo disorder in $Y_{0.8}U_{0.2}Pd_3$ cannot be completely ruled out at this time.

To date μSR studies of $Y_{1-x}U_xPd_3$ have been carried out only in zero and longitudinal applied field [16]. Transverse-field μSR measurements are currently under way to complement the NMR results.

9. Summary

We have seen that magnetic resonance experiments (NMR and μSR) can provide direct evidence for a wide distribution of Kondo temperatures as a mechanism for non-Fermi-liquid behaviour in heavy-fermion alloys. In the non-Fermi-liquid alloy system $UCu_{5-x}Pd_x$ NMR and μSR linewidths are large and can be explained by a simple Kondo-disorder model. The good agreement between μSR and NMR results in the limit of short-range correlation of the inhomogeneous susceptibility is evidence against the possibility that macroscopic inhomogeneity (chemical clustering, phase segregation, ...) plays a major role; the Kondo-disorder behaviour appears to be intrinsic. The Kondo-disorder picture is able to explain many of the NFL properties, but it is not yet clear whether all NFL behaviour can be accounted for in detail in this model. Work is under way to address these issues.

In $CeCu_{5.9}Au_{0.1}$ only μSR experiments have been carried out to date. The SRC limit also seems most likely to be applicable to this system, but a conclusive test will require NMR data. Kondo disorder may be present, but is not dominant if the SRC limit is applicable. This accords with the very convincing evidence [24] for quantum critical phenomena as the origin of NFL behaviour in this alloy.

NMR in $Y_{1-x}U_xPd_3$ shows a change of lineshape for $T \lesssim T_K$ but no superlinear increase of the linewidth dependence on χ as required by the Kondo-disorder mechanism. The origin of the observed NMR behaviour is not well understood, but there is no clear sign of Kondo disorder.

We conclude that Kondo disorder is an important mechanism in at least one NFL alloy system ($UCu_{5-x}Pd_x$); the search is on for others. Clearly not all NFL systems exhibit Kondo disorder, which is certainly not the only possible origin of NFL behaviour. But only a local probe such as magnetic resonance can determine the importance of Kondo disorder in any given NFL system.

Acknowledgments

It is a pleasure to acknowledge our collaborators, without whom this work would have not been possible: L P Le and R H Heffner, Los Alamos National Laboratory; A Amato, R Feyerherm, F N Gygax, H R Ott and A Schenck, ETH Zurich; G J Nieuwenhuys, Rijksuniversiteit Leiden; B Andraka, University of Florida; H von Löhneysen and O Stockert, Universität Karlsruhe; and C L Seaman and M B Maple, University of California, San Diego. This research was supported by the US National Science Foundation, Grant No DMR-9418991, and by the University of California, Riverside, Academic Senate Committee on Research.

Appendix A. The relationship between the susceptibility distribution width and spin-probe linewidth

We assume for the moment that the a_{ij} in equation (4) are the same for crystallographically equivalent values of i and j; disorder in a_{ij} is treated in appendix B. From equation (4) we construct the spatial averages

$$\langle K\rangle = a\langle\chi\rangle \qquad a \equiv \sum_j a_{ij}$$

and

$$\kappa^2 \equiv \langle\delta K^2\rangle = \sum_{jk} a_{ij}a_{ik}\langle\delta\chi_j\,\delta\chi_k\rangle. \tag{A1}$$

Here $\delta K = K - \langle K\rangle$ and $\delta\chi \equiv \chi - \langle\chi\rangle$ are the deviations of the shift and susceptibility, respectively, from their averages. Thus the rms width κ depends on the correlation function $\langle\delta\chi_j\,\delta\chi_k\rangle$, and in particular on the correlation length ξ which characterizes it.

In the limit of long-range correlation (LRC) the susceptibility is disordered in macroscopic domains, i.e., ξ is much longer than the local-moment near-neighbour spacing. Then each spin probe senses a locally uniform susceptibility, so $\langle\delta\chi_j\,\delta\chi_k\rangle \approx \langle\delta\chi^2\rangle$ may be factored out of the summation in equation (A1). This yields

$$\kappa^2 = a^2\langle\delta\chi^2\rangle \qquad \text{(LRC)}$$

and so

$$\kappa = |a|(\delta\chi)_{\rm rms} \qquad \text{(LRC)}$$

and the fractional width $(\delta\chi)_{\rm rms}/\langle\chi\rangle$ is

$$(\delta\chi)_{\rm rms}/\langle\chi\rangle = \kappa/(a^*\langle\chi\rangle) \tag{A2}$$

with

$$a^* = a^*_{\rm LRC} = |a| = \left|\sum_j a_{ij}\right|. \tag{A3}$$

This limit was implicitly assumed in reference [14] but, as discussed in the text, is incorrect in the case of $UCu_{5-x}Pd_x$.

In the extreme opposite limit of short-range correlation (SRC) ξ is much shorter than the moment spacing, i.e., the variation of χ_j from site to site is random [28]. Then

$$\langle\delta\chi_j\,\delta\chi_k\rangle = \begin{cases}\langle\delta\chi^2\rangle & j = k \qquad \text{(SRC)}\\ 0 & j \neq k \qquad \text{(SRC)}.\end{cases}$$

In this case one finds

$$\kappa^2 = \sum_j a_{ij}^2\langle\delta\chi^2\rangle \qquad \text{(SRC)}$$

so that again $(\delta\chi)_{\rm rms}/\langle\chi\rangle = \kappa/(a^*\langle\chi\rangle)$, but with

$$a^* = a^*_{\rm SRC} = \left(\sum_j a_{ij}^2\right)^{1/2}. \tag{A4}$$

Appendix B. The spin-probe linewidth due to disorder in both the susceptibility and the coupling constants

We now consider the case where both χ and the a_{ij} are disordered. Then

$$\langle K\rangle = \sum_j \langle a_{ij}\chi_j\rangle \qquad \text{and} \qquad \langle K^2\rangle = \sum_{jk}\langle a_{ij}a_{ik}\chi_j\chi_k\rangle.$$

It is difficult to go further without assuming that the distributions of a_{ij} and χ are uncorrelated. Making this assumption, one finds that

$$\langle K\rangle = \langle a\rangle\langle\chi\rangle \qquad \langle a\rangle = \sum_j\langle a_{ij}\rangle$$

and

$$\kappa^2 = \sum_{jk}(\langle\delta a_{ij}\,\delta a_{ik}\rangle\langle\chi\rangle^2 + \langle a_{ij}a_{ik}\rangle\langle\delta\chi_j\,\delta\chi_k\rangle). \tag{B1}$$

Thus we have a term in $\langle\chi\rangle^2$, with a coefficient proportional to the deviations of the coupling constants, in addition to the original term in $\langle\delta\chi_j\,\delta\chi_k\rangle$.

Specializing to the SRC limit for both the χ_j and the a_{ij}, equation (B1) gives

$$\kappa^2 = \langle\delta a^2\rangle\langle\chi\rangle^2 + \langle a^2\rangle\langle\delta\chi^2\rangle \tag{B2}$$

so the two contributions to κ^2 are both positive.

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