

Understanding the origin of non-Fermi liquid behaviour in doped Kondo insulators

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Abstract

We have made inelastic neutron scattering measurements on the doped Kondo insulators $\text{Ce}(\text{Rh}_{0.8}\text{Pd}_{0.2})\text{Sb}$ and $(\text{Ce}_{0.7}\text{Th}_{0.3})\text{RhSb}$. Previous bulk property measurements showed non-Fermi liquid (NFL) behaviour for both compositions. In our inelastic neutron scattering data these two compounds also show E/T scaling behaviour in the dynamical spin susceptibility with different exponents, which is considered a characteristic feature of NFL systems. We discuss how these two different exponents might be related to different microscopic mechanisms of the NFL behaviour in the bulk properties as well as the E/T scaling behaviour seen in the dynamical susceptibility.

1. Introduction

Theories based on the Fermi liquid (FL) quasiparticle concept have been very successful over the last 40 years in describing the low-temperature properties of metals, including strongly correlated electron systems. The main predictions of these theories are that well below the characteristic Fermi temperature, T_F : (1) the resistivity should exhibit a quadratic temperature dependence, (2) the heat capacity should show a linear temperature dependence, and (3) the susceptibility should be temperature independent. These predictions have been borne out in many, many metals and seemed to allow no exceptions until recent experiments demonstrated that such exceptions do indeed exist in some heavy-fermion compounds [1]. These systems, growing in number, show physical properties significantly deviating from the predictions of

FL theories at low temperatures: $\rho \propto T^\eta$ ($1 \leq \eta \leq 2$), $C_v \propto T \ln(T_0/T)$ or $\gamma_0 - T^{1/2}$, and $\chi \propto \ln T$, i.e. so-called non-Fermi liquid (NFL) behaviour.

Another interesting feature of the NFL systems is that they show unusual E/T scaling behaviour in the dynamical spin susceptibility, $\chi''(\omega, T)$, measured using the inelastic neutron scattering technique. This then implies that the temperature (T) itself is the energy scale governing the spin fluctuations. This E/T scaling behaviour has been so far observed in $\text{U}(\text{Cu}, \text{Pd})_5$ by Aronson *et al* [2] and $\text{Ce}(\text{Cu}, \text{Au})_6$ by Schroder *et al* [3]. However, we note that the two groups' experimental results produce different scaling exponents in the dynamical susceptibility: 0.33 for the former and 0.77 for the latter. This difference in scaling exponent, we believe, is deeply related to the underlying microscopic mechanism of NFL behaviour. Theoretical scenarios proposed for the NFL behaviour include the $T = 0$ quantum phase transition theory (QPT), the two-channel Kondo model, and the Kondo disorder model.

CeRhSb is a Kondo insulator with a small energy gap ($\Delta \simeq 4$ K) at E_F . With Pd doping on the Rh site: $\text{Ce}(\text{Rh}_{1-x}\text{Pd}_x)\text{Sb}$, the gap is rapidly suppressed and at $x = 0.2$ it exhibits NFL behaviour in the heat capacity, $C \propto T \ln T$ [4]. Furthermore, a new antiferromagnetic state develops for higher Pd concentration up to $x = 0.4$. A very similar phase diagram was also observed for Th-doped CeRhSb , where NFL behaviour is found for 30% Th doping [5].

2. Experimental details

For our experiments, we used 30 g of polycrystalline $\text{Ce}(\text{Rh}_{0.8}\text{Pd}_{0.2})\text{Sb}$ and 18 g of $(\text{Ce}_{0.7}\text{Th}_{0.3})\text{RhSb}$ prepared in small quantities (5–7 g) by arc-melting high-purity elements. To ensure their homogeneity, the samples were flipped and melted 3–5 times. Inelastic neutron scattering measurements were made with the HET chopper spectrometer at the UK ISIS spallation neutron source using incident energies of 23, 60, 80, and 500 meV. We also measured lower-energy spectra of $\text{Ce}(\text{Rh}_{0.8}\text{Pd}_{0.2})\text{Sb}$ with an incident energy of 3.1 meV using the time-of-flight spectrometer IN6 at the ILL, Grenoble. We made measurements on LaRhSb , which has the same crystal structure as CeRhSb , as a phonon-blank sample. All our data have been corrected for phonon contributions by following a conventional phonon subtraction procedure. Details of our experiments are given elsewhere [6].

3. Results and discussion

In the raw data for both compositions, we found that the data on the energy loss side collapse on top of each other below about 10 meV while the spectra are strongly temperature dependent on the energy gain side. We note that all the data satisfy the detailed balance principle. What is particularly interesting is that the data in the energy range of 0.1–10 meV exhibit a scaling behaviour for the magnetic scattering, $S_{mag}(\omega, T) \propto \omega^{-\alpha}$ with $\alpha = 0.77$ for $\text{Ce}(\text{Rh}_{0.8}\text{Pd}_{0.2})\text{Sb}$ [6], and a similar scaling behaviour is also found with $\alpha = 0.33$ for $(\text{Ce}_{0.7}\text{Th}_{0.3})\text{RhSb}$ over a narrower energy range of 1 to 10 meV [7]. On the other hand, we did not observe the scaling behaviour in either $\text{Ce}(\text{Rh}_{0.9}\text{Pd}_{0.1})\text{Sb}$ or $\text{Ce}(\text{Rh}_{0.7}\text{Pd}_{0.3})\text{Sb}$. This kind of scaling behaviour is most clearly seen in the dynamical susceptibility ($\chi''(\omega, T)$) obtained from the scattering function. Figures 1(a) and (b) show E/T scaling of $\chi''(\omega, T)$ for both compounds using the aforementioned values of α . We were able to fit the E/T scaling behaviour using the function which was previously employed for the analysis of $\text{U}(\text{Cu}, \text{Pd})_5$ [2]. That we have observed two different scaling exponents is a strong indicator that the NFL behaviour in the two compositions arises from different mechanisms.

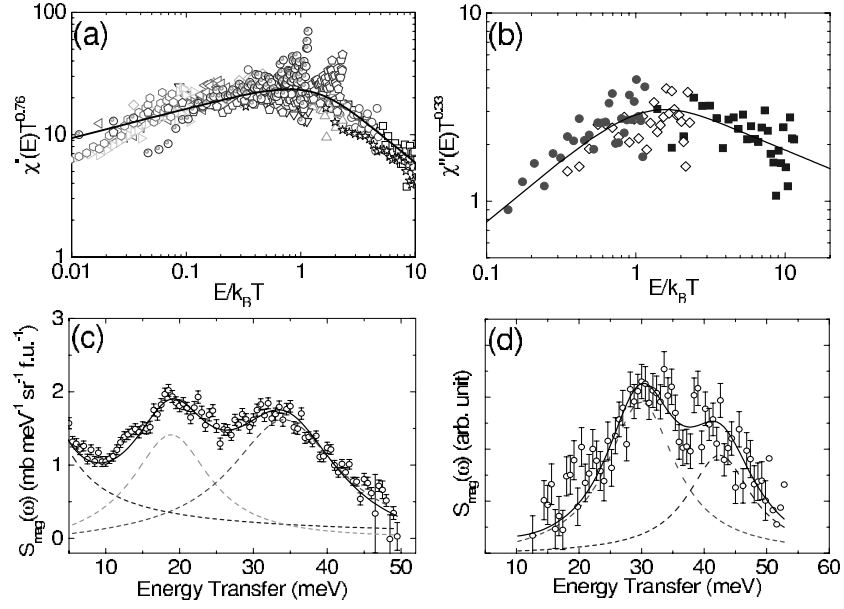


Figure 1. $\chi''T^\alpha$ for (a) $\text{Ce}(\text{Rh}_{0.8}\text{Pd}_{0.2})\text{Sb}$ with $\alpha = 0.77$ and (b) $(\text{Ce}_{0.7}\text{Th}_{0.3})\text{RhSb}$ with $\alpha = 0.33$; and magnetic scattering from localized Ce moments of (c) $\text{Ce}(\text{Rh}_{0.8}\text{Pd}_{0.2})\text{Sb}$ and (d) $(\text{Ce}_{0.7}\text{Th}_{0.3})\text{RhSb}$. The curves in (a) and (b) represent our curve fitting results obtained using the function given [2], while the curves in (c) and (d) are curve fitting results obtained using the CEF Hamiltonian (see the text).

A further interesting point is the presence of crystalline electric field (CEF) excitations above 10 meV in our data. As shown in figures 1(c) and (d), two well-defined CEF excitations are observed for $\text{Ce}(\text{Rh}_{0.8}\text{Pd}_{0.2})\text{Sb}$ while much broader CEF excitations are present in $(\text{Ce}_{0.7}\text{Th}_{0.3})\text{RhSb}$. In order to analyse the data, we used the following CEF Hamiltonian for localized Ce moments with the a -axis as their quantization axis: $H_{\text{CEF}} = B_{20}O_{20} + B_{40}O_{40} + B_{43}O_{43}$. In this Hamiltonian, Ce 4f levels are split into three doublets with two doublet states forming out of a mixture of $|\pm 1/2\rangle$ and $|\mp 5/2\rangle$, and one doublet state with pure $|\pm 3/2\rangle$. The curves through the data points in figures 1(c) and (d) are results of our calculation using the Hamiltonian with the ground state being pure $|\pm 3/2\rangle$. It is to be noted that similarly good agreement can also be found with the ground state of a mixture of $|\pm 1/2\rangle$ and $|\mp 5/2\rangle$ and pure $|\pm 3/2\rangle$ being the first excited state. We also note that similar CEF excitations have been observed for $\text{Ce}(\text{Rh}_{0.9}\text{Pd}_{0.1})\text{Sb}$, $\text{Ce}(\text{Rh}_{0.7}\text{Pd}_{0.3})\text{Sb}$, and $\text{Ce}(\text{Ni}_{0.935}\text{Cu}_{0.065})\text{Sn}$ [8], which can be fitted using the same Hamiltonian with almost the same CEF level schemes as above. The observation of CEF levels in both $\text{Ce}(\text{Rh}_{0.8}\text{Pd}_{0.2})\text{Sb}$ and $(\text{Ce}_{0.7}\text{Th}_{0.3})\text{RhSb}$ indicates that the NFL behaviour in these systems is confined to an energy scale below about 10 meV and that they exhibit a more localized behaviour for energies above it. It is to be noted that this energy scale corresponds well with the characteristic energy scale obtained from the bulk properties.

CEF excitations are usually sensitive to the strength of the hybridization J between localized f electrons and conduction electrons. For materials with larger J such as mixed-valence and heavy-fermion compounds, the excitations are usually very broad and not easily observable from inelastic neutron scattering experiments. Therefore, that we have observed CEF excitations in $\text{Ce}(\text{Rh}_{0.8}\text{Pd}_{0.2})\text{Sb}$ and $(\text{Ce}_{0.7}\text{Th}_{0.3})\text{RhSb}$ strongly indicates that

this hybridization J is reduced with doping either Pd on the Rh sites, or Th on the Ce sites. However, the different degrees of sharpness of the CEF excitations for the two compositions imply that Pd doping is more effective in reducing J -values than Th doping.

Although several groups have so far observed the E/T scaling behaviour in some heavy-fermion compounds showing NFL behaviour in the bulk properties, unfortunately it is not well established at the moment whether the different exponents obtained in the dynamical susceptibility plots are entirely due to different microscopic mechanisms specific to each system. In fact, one of the main motivations behind our systematic studies is to find out from experiments whether this is true. In our case, despite $\text{Ce}(\text{Rh}_{0.8}\text{Pd}_{0.2})\text{Sb}$ and $(\text{Ce}_{0.7}\text{Th}_{0.3})\text{RhSb}$ having different degrees of hybridization (J), they show similar E/T scaling behaviours only with different exponents. This indicates that there ought to be another factor at work here related to the E/T scaling behaviour. As one of the possibilities, we consider that different doping sites may be a relevant issue here. For example, Pd doping on the ligand sites does not disturb the Ce lattice whereas Th doping disrupts the Ce lattice. If the different exponents are indeed due to different microscopic mechanisms, then this difference in the doping effects is more likely to be the reason for that. As we discussed in our previous publication [6], there are three major routes to NFL behaviour: the $T = 0$ quantum phase transition scenario, the two-channel Kondo model, and the Kondo disorder model. However, the two-channel Kondo model cannot work for Ce compounds since there is no known Ce compound satisfying the criterion of the two-channel Kondo model. Furthermore, we found in the previous work that the E/T scaling behaviour and thus the NFL features found in $\text{Ce}(\text{Rh}_{0.8}\text{Pd}_{0.2})\text{Sb}$ are more likely to be due to the $T = 0$ quantum phase transition. Therefore, we are left with only one viable explanation for the E/T scaling behaviour found in $(\text{Ce}_{0.7}\text{Th}_{0.3})\text{RhSb}$. That NFL behaviour in the heat capacity of $(\text{Ce}_{1-x}\text{Th}_x)\text{RhSb}$ seems to be found in a broad x -range between 0.2 and 0.4 [5] may also be in agreement with our conclusion that the Kondo disorder model is more appropriate for the unusual behaviour of $(\text{Ce}_{0.7}\text{Th}_{0.3})\text{RhSb}$. This doping dependence is again different from what we found in our inelastic neutron studies of several $\text{Ce}(\text{Rh}_{1-x}\text{Pd}_x)\text{Sb}$ compounds with $x = 0, 0.1, 0.2,$ and 0.3 , in which the E/T scaling behaviour is only seen for the $x = 0.2$ sample [6]. However, we acknowledge that this conclusion needs to be supported by further measurements and analysis.

4. Summary

In summary, we have observed clear E/T scaling behaviour in the dynamical susceptibility of $\text{Ce}(\text{Rh}_{0.8}\text{Pd}_{0.2})\text{Sb}$ and $(\text{Ce}_{0.7}\text{Th}_{0.3})\text{RhSb}$. We also implied that the measurements of CEF excitations together with considerations of the bulk properties, in particular the doping dependence, can be useful in determining the origin of the E/T scaling behaviour. On the basis of our measurements, we think that the NFL behaviour in $\text{Ce}(\text{Rh}_{0.8}\text{Pd}_{0.2})\text{Sb}$ originates from the $T = 0$ QCP whilst the NFL behaviour in $(\text{Ce}_{0.7}\text{Th}_{0.3})\text{RhSb}$ may be due to the Kondo disorder.

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