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Non-Fermi liquid behaviour in the dynamic susceptibility of $Ce(Rh_{0.8}Pd_{0.2})Sb$

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Abstract

We have made inelastic neutron scattering studies of $Ce(Rh_{0.8}Pd_{0.2})Sb$, which is located at the crossover from non-magnetic to magnetically ordered states (i.e. close to a quantum critical point). In the imaginary part of the dynamic susceptibility, we have found clear evidence of E/T scaling behaviour. This E/T scaling behaviour appears to break down at high temperatures and energies. The transition from non-Fermi liquid to localized behaviour in $Ce(Rh_{0.8}Pd_{0.2})Sb$ on an energy scale above $\sim$15 meV is demonstrated by the observation of both crystal-field and spin–orbit excitations of the Ce^{3+} ions.

1. Introduction

Over the last two decades or so, heavy-fermion compounds have been studied extensively and intensively [1]. Only recently, however, has it been observed that under certain conditions some heavy-fermion systems, for example $U_xY_{1-x}Pd_3$ [2] and $CeCu_{6-x}Au_x$ [3], display transport and thermodynamic properties that deviate significantly from what one would expect within the Fermi liquid description. This so-called non-Fermi liquid (NFL) behaviour has since been observed in many other heavy-fermion systems [4].

Although the physical properties of the NFL systems deviate from the predictions of the conventional Fermi liquid picture, their actual temperature dependence varies from one material to another. This difference is presumably related to different microscopic origins of the NFL behaviour. For example, the low-temperature resistivity of $(U_{0.2}Y_{0.8})Pd_3$, one of the first heavy-fermion compounds shown to exhibit NFL behaviour, follows a $-\ln T$ behaviour and the susceptibility $T^{-\eta}$ with $\eta = 0.3$ [2]. Heat capacity data for $(U_{0.2}Y_{0.8})Pd_3$ also show an unusual temperature dependence of $-T \ln T$. On the other hand, the resistivity

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is a linear function of temperature below 1 K in $Ce(Cu_{5.9}Au_{0.1})$ while the heat capacity shows $-T\ln(T/T_o)$ dependence [3], where T_o is the characteristic temperature.

Another interesting feature of NFL heavy-fermion compounds is that they show an unusual E/T scaling behaviour in their dynamic susceptibility χ'' measured from inelastic neutron scattering experiments. This E/T scaling behaviour has been observed so far in $U(Cu, Pd)_5$ [5] and $Ce(Cu, Au)_6$ [6] with different exponents. Interestingly, the E/T scaling functions exhibit different behaviours in the two compounds. Undoubtedly, this E/T scaling behaviour is deeply related to the underlying microscopic mechanism of each material. Nonetheless, our understanding of the E/T scaling observed in χ'' is largely lacking.

As regards the origin of the NFL behaviour, a few theoretical scenarios have been proposed so far. One is the two-channel Kondo model, which is thermodynamically equivalent to the quadrupolar Kondo model [7]. This model was originally proposed to explain the NFL behaviour of $(U_{0.2}Y_{0.8})Pd_3$. However, this interpretation has been questioned by several authors after new experiments including a recent inelastic neutron study [8] suggested that the ground state of $(U_{0.2}Y_{0.8})Pd_3$ should be a mixture of non-magnetic Γ_3 and magnetic Γ_5, not pure Γ_3 as required in the two-channel Kondo model. Furthermore, our recent resistivity measurements on $(U_xY_{1-x})Pd_3$ with $x = 0.2$ and 0.45 suggest that the NFL behaviour is more likely to be due to metallurgical disorder in the chemical composition [9]. Another route to the NFL behaviour is the Kondo disorder model, which shows that a distribution of Kondo temperatures can lead to unconventional low-temperature properties [10, 11]. This Kondo disorder model seems to be appropriate for the NFL behaviour of $U(Cu_{5-x}Pd_x)$ with $x = 1$ and 1.5, as shown by the NMR study [10]. Lastly, there is the $T = 0$ quantum phase transition scenario [12]. According to this theory, when a magnetic transition is suppressed toward $T = 0$ K with pressure, magnetic field, or alloying, the physical properties at this critical point are fundamentally different from the conventional FL predictions. This $T = 0$ quantum phase transition theory seems to be very successful at explaining the NFL behaviours of $Ce(Cu, X)_6$ with X = Ag and Au [3, 13]. A newly proposed theoretical model invoking the Griffiths phase [14] has ingredients of both the Kondo disorder model and the $T = 0$ quantum phase transition. Given the intricate nature of the theories as well as the experimental results, it has been a very challenging and difficult task to determine the criteria for the choice of the appropriate model for specific systems.

CeRhSb is one of the family of low-carrier-density Kondo systems with a very small gap energy of $E_g = 4$ K [15]. With Pd doping on the Rh site, the gap is rapidly suppressed and expected to disappear at 20% Pd doping [16]. For larger Pd doping, a new antiferromagnetic state develops until 40% doping of Pd is reached. At the critical doping of 20% Pd, there is a crossover from non-magnetic to magnetically ordered states with a $T\ln T$ behaviour in the heat capacity [16], similar to the NFL behaviour in other systems.

In the present study, we have investigated the dynamic susceptibility (χ'') of $Ce(Rh_{0.8}Pd_{0.2})Sb$ using inelastic neutron scattering, where we found a clear E/T scaling behaviour. Unlike those from the two previous studies on $U(Cu, Pd)_5$ and $Ce(Cu, Au)_6$, our results for $Ce(Rh_{0.8}Pd_{0.2})Sb$ shows a clear crossover from a NFL regime to a classical regime with increasing temperatures and energies that has not been reported before. We discuss our results with reference to the three theoretical scenarios, finding that the E/T scaling behaviour of $Ce(Rh_{0.8}Pd_{0.2})Sb$ is more consistent with the $T = 0$ quantum phase transition theory.

2. Experimental details

About 30 g of polycrystalline $Ce(Rh_{0.8}Pd_{0.2})Sb$ and LaRhSb samples were 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 have been 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 with an incident energy of 3.1 meV using the time-of-flight spectrometer IN6 at the ILL, Grenoble.

The scattering function $S(Q, \omega)$ of our HET data is given in absolute units of mb meV^{-1} sr^{-1} fu^{-1} as a function of wavevector transfer, Q, and energy transfer, $\hbar\omega$, having been obtained from the raw time-of-flight data by normalizing to the incoherent scattering from a flat-vanadium-slab standard sample for each chosen incident energy, and then to the incident flux using the integral of the incident beam monitor. The low-angle HET data were corrected for phonon contributions by scaling the high-angle scattering ($\phi = 133^\circ$) to the low-angle scattering ($\phi = 9.3^\circ \rightarrow 28.7^\circ$) and using the standard scaling method with a scaling factor, obtained by making measurements on the LaRhSb sample with the same incident energy. On the other hand, for the IN6 data we made measurements on LaRhSb in the same conditions as for $Ce(Rh_{0.8}Pd_{0.2})Sb$, and found that there is very little phonon contribution below 2 meV; hence we used the total measured scattering in our analysis. Since Rh is a strong neutron absorber, we have made absorption corrections to our inelastic scattering data using a FORTRAN program written by Perring at ISIS.

3. Results and discussion

In figure 1, we show the total inelastic spectra taken from IN6 (ILL) together with the inelastic spectra with phonon subtraction taken from HET (ISIS). Our HET data were measured with an incident energy of 23 meV. In order to present the IN6 data in terms of absolute units, we have multiplied the IN6 data by a factor to scale them to the same value as the HET data at 100 K at an energy transfer of 1.5 meV. As one can see, it is noticeable that all the data on the energy-loss side collapse on top of each other, while the spectra are strongly temperature dependent on the energy-gain side. We have checked whether the spectra obey the detailed balance principle by calculating the spectra on the energy-gain side using the relation of $S(\omega > 0)\exp(-\omega/T) = S(\omega < 0)$ for the IN6 data. As one can see from the figure, our calculations reproduce the spectra on the energy-gain side rather well.

A double-logarithmic plot of the magnetic scattering function against energy transfer is particularly interesting. As shown in figure 2, clearly all the spectra in the energy range of 0.1–10 meV show a scaling behaviour of $S(\omega) \approx \omega^{-0.77}$. We note that the normalization of the IN6 data does not affect the scaling behaviour at all since any multiplication only shifts the data upwards or downwards in the log–log plot. A very similar scaling behaviour of $S(\omega)$ has been previously observed in $U(Cu_{5-x}Pd_x)$ [5], but with an exponent of $1/3$ instead of 0.77. It is also noticeable that the data points taken at $T \geqslant 200$ K suggest that there appears a systematic deviation, marked by a dotted ellipse, in the scaling behaviour of $S_{mag}(\omega)$ at high temperatures.

The imaginary part of the dynamic susceptibility $\chi''(\omega, T)$ and the scattering function $S(\omega)$ are related by

$$S(\omega) = \frac{1}{1 - \exp(-\hbar\omega/k_B T)}\chi''(\omega, T). \tag{1}$$

In the plot of $\chi''(\omega, T)$ for different temperatures (figure 3), we observed that $\chi''(\omega, T)$ showed different scaling behaviours depending on whether $\hbar\omega > k_B T$ or $\hbar\omega < k_B T$. This suggests that $\chi''(\omega, T)$ should have a specific function. Incidentally, a similar behaviour has been previously seen in $\chi''(\omega, T)$ for $U(Cu_{5-x}Pd_x)$ by Aronson *et al* [5]. In their paper, they inferred that the specific $\chi''(\omega, T)$ behaviour required a certain ω/T dependence. Following

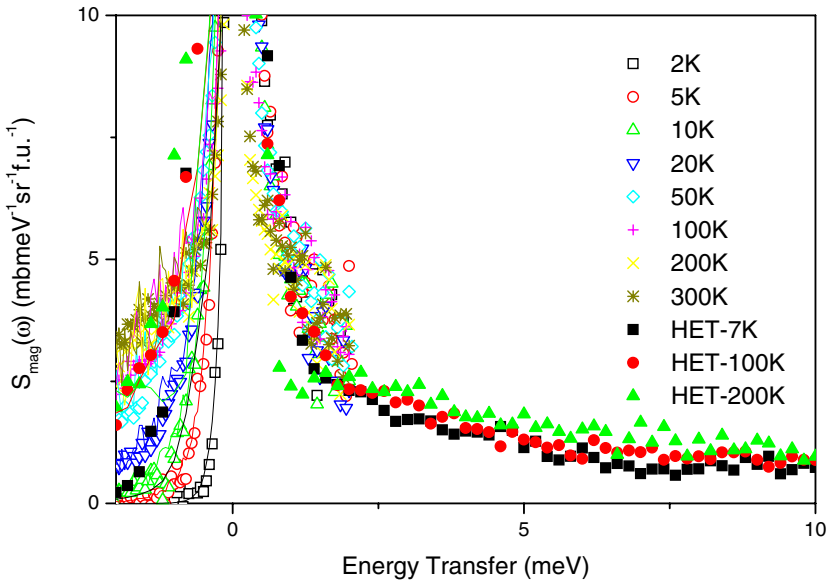

Figure 1. The magnetic scattering function $S_{mag}(\omega)$ measured on IN6 (ILL) and HET (ISIS) spectrometers for several temperatures. The curves through the IN6 data (unfilled symbols) are results calculated from the data on the energy-loss part using the detailed balance factor.

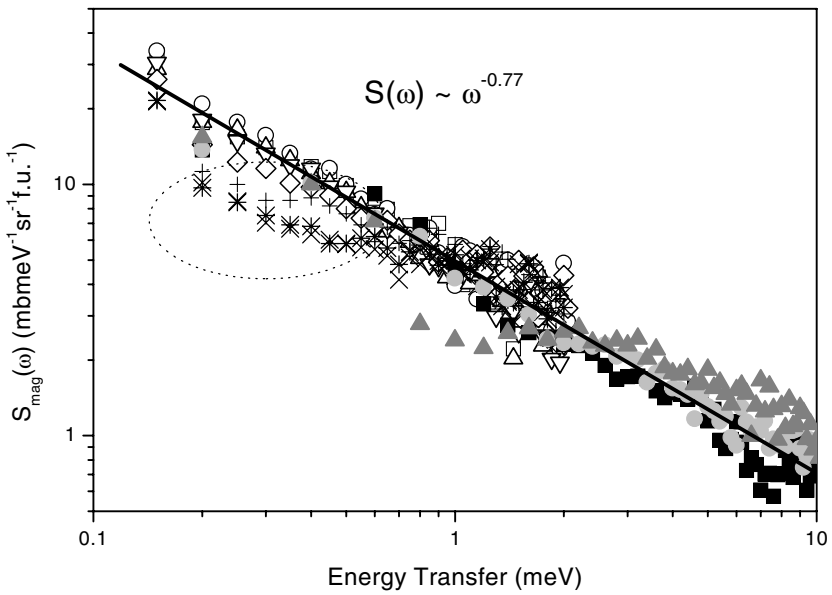

Figure 2. A log–log plot of both the IN6 (open symbols) and the HET data (filled symbols) on the energy-loss part. The solid line shows $S_{mag}(\omega) \sim \omega^{-0.77}$ scaling behaviour. The dotted ellipse indicates a region where the scaling behaviour breaks down at high temperatures (see the text). We used the same symbols for each temperature as in figure 1.

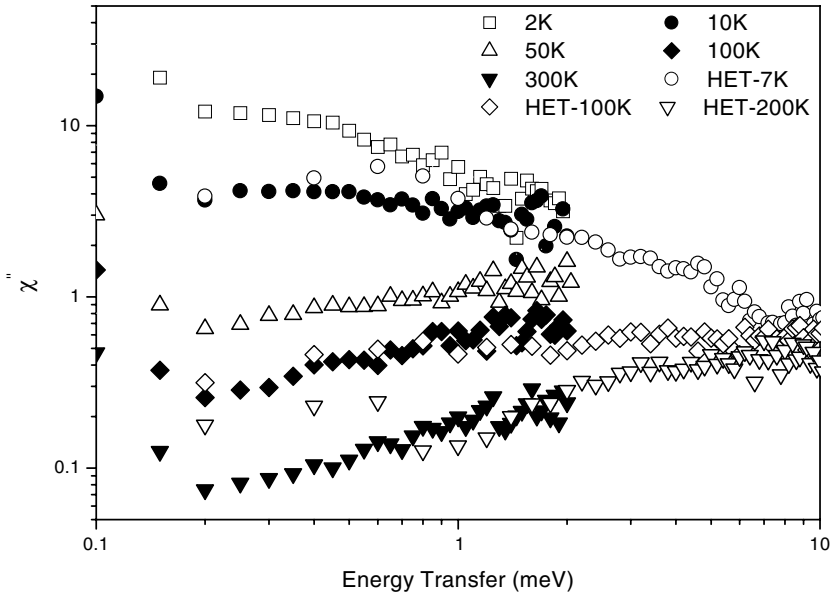

Figure 3. χ'' versus E for several temperatures. For the sake of clarity, we do not show all the data (see the text).

Aronson's treatments of $\chi''(\omega, T)$, we find that our $\chi''(\omega, T)$ for $Ce(Rh_{0.8}Pd_{0.2})Sb$ can be best described by

$$\chi''(\omega, T)T^{\alpha} \simeq Z(\omega/T) \tag{2}$$

with $\alpha = 0.77$. Again we have a different exponent (α) value from that found in $\chi''(\omega, T)$ for $U(Cu_{5-x}Pd_x)$ [5]. As shown in figure 4, there is a clear universal behaviour in the plot of equation (2). In order to analyse the data plotted in figure 4, we have used firstly a function given in [5]:

$$Z(\omega, T) \simeq (T/\omega)^{\alpha} \tanh(\omega/\beta T) \tag{3}$$

with $\alpha = 0.77$ and $\beta = 1.2$. The solid line displays the results that we obtained. From our analysis, we find that equation (3) fits the experimental data reasonably well for β-values in the range of $1 \leqslant \beta \leqslant 1.5$. However, the agreement is strongly sensitive to the α-value chosen. For example, the dashed line shows the function obtained with $\alpha = 1/3$ and $\beta = 1.2$. Clearly it does not describe the data well. We also considered the function employed by Schroder *et al* for $Ce(Cu_{6-x}Au_x)$ [6]:

$$Z(\omega/T) \simeq \sin[\alpha \tan^{-1}(\omega/T)]/[(\omega/T)^2 + 1]^{\alpha/2}. \tag{4}$$

The dash–dot curve shows the results obtained with $\alpha = 0.77$. Although it describes the data in the regime of $E/k_BT > 1$, it falls more rapidly than the data points in the region of $E/k_BT < 1$.

With increasing energies, however, the E/T scaling behaviour breaks down and the system becomes localized. In figures 5(a) and (b), we show the magnetic scattering $S_{mag}(\omega)$ and the total scattering from $Ce(Rh_{0.8}Pd_{0.2})Sb$ measured on the HET spectrometer with incident energies of 80 and 500 meV, respectively. As one can see in figure 5(a), there are two clear excitations centred at 19 and 34 meV, respectively. These are attributed to the crystalline-electric-field (CEF) excitations of the $J = 5/2$ ground states of Ce^{3+}. For comparison, we

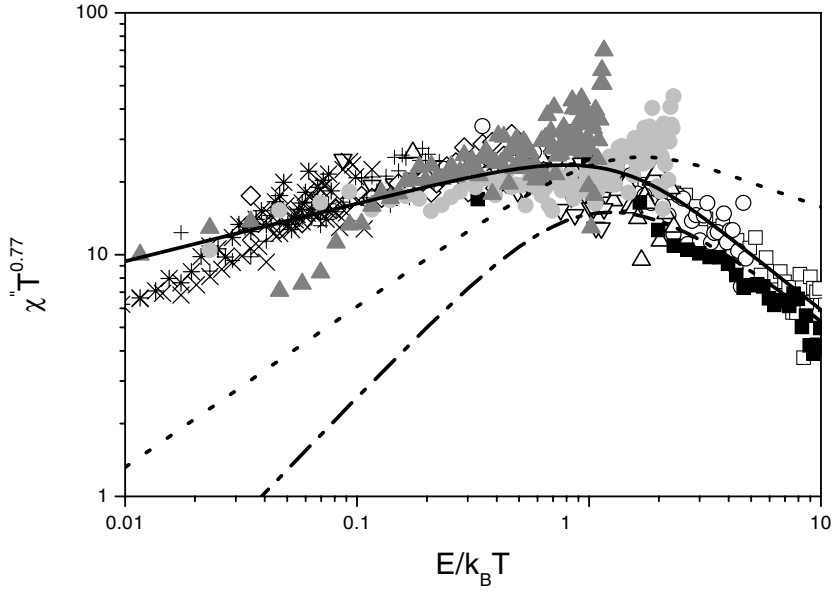

Figure 4. A $\chi''T^{0.77}$ versus E/T plot. The solid curve is for equation (3) with $\alpha = 0.77$, the dashed curve for equation (3) with $\alpha = 1/3$ for U($Cu_{5-x}Pd_x$) [5], and the dash–dot curve for equation (4) for Ce($Cu_{6-x}Au_x$) [6]. We used the same symbols for each temperature as in figure 1.

also show magnetic scattering from CeRhSb that is almost featureless over the energy range. Interestingly enough, two CEF excitations are also observed in Ce(Ni, Cu)Sn that become extremely broadened for CeNiSn [17]. The presence of the two clear CEF excitations in figure 5(*a*) indicates that the Ce^{3+} ions of Ce($Rh_{0.8}Pd_{0.2}$)Sb have a substantial amount of localized 4f character. Further evidence of the 4f electron localization is seen in figure 5(*b*), which shows a magnetic excitation centred at 286 meV, which we attribute to the $^2F_{5/2} \rightarrow {}^2F_{7/2}$ spin–orbit (SO) transition. A similar SO transition was observed at 260 and 291 meV for $CeAl_3$ [18]. Therefore, our observation of both CEF and SO excitations suggests that the Ce ions of Ce($Rh_{0.8}Pd_{0.2}$)Sb behave as a localized system above $E \simeq 15$ meV.

Interestingly, we note that there is a sign of breakdown of the scaling behaviour (marked by a dotted ellipse) in figure 2 above 200 K, corresponding to about 15 meV. Thus there is a crossover from the NFL regime to a normal localized regime for high temperatures and high energies. This is the first time that such a crossover from the NFL to the localized regime, with increasing temperatures and energies, has been so clearly demonstrated.

In order to shed light on the microscopic origin of the NFL behaviour, in particular its possible antiferromagnetic nature, we studied the wavevector dependence of the magnetic scattering at low energies. Firstly, we note that Ce($Rh_{0.8}Pd_{0.2}$)Sb exhibits quasielastic magnetic scattering (centred around zero energy transfer), which was absent in the parent Kondo insulator compound CeRhSb [19]. The presence of quasielastic scattering and well defined crystal-field excitations in Ce($Rh_{0.8}Pd_{0.2}$)Sb are consistent with the bulk measurements, which reveal the absence of an energy gap at the Fermi energy. To estimate the wavevector dependence of the quasielastic linewidth we have analysed the 5 K data from IN6. The observed 235 spectra from IN6 were grouped into ten Q-groups ($Q = 0.32$–2.02 Å^{-1}). The quasielastic linewidth was estimated by fitting the individual grouped spectra to a quasielastic Lorentzian function and instrument resolution function, the latter being a convolution of Gaussian and Lorentzian

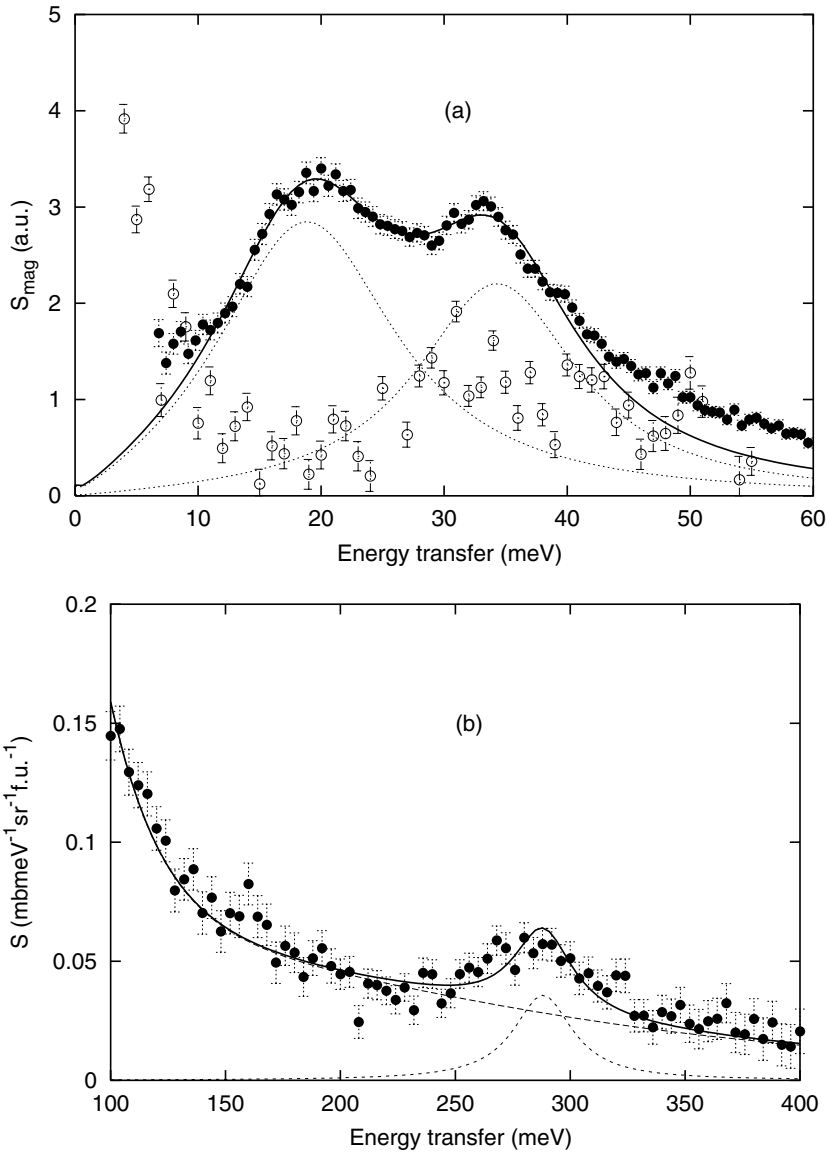

Figure 5. (*a*) The magnetic scattering function (closed circles) $S_{mag}(\omega)$ taken with an incident energy of 80 meV at 7 K. Two Lorenztians centred at 19 and 34 meV are used to describe the CEF excitations. For comparison, we show magnetic scattering from pure CeRhSb (open circles) taken from [19]. (*b*) The high-energy part of the scattering function taken with an incident energy of 500 meV. The peak at 286 meV is for the $^2F_{5/2} \rightarrow {}^2F_{7/2}$ SO transition.

functions. The width and relative intensity parameters of the instrument resolution functions were estimated from a vanadium run with identical conditions and were kept fixed in the fitting. Figure 6 shows the half-width, Γ, as a function of q. From the data one can see that the quasielastic linewidth is almost q-independent. This is a characteristic of single-ion Kondo behaviour and suggests that the intersite RKKY coupling is suppressed by the Kondo effect in $Ce(Rh_{0.8}Pd_{0.2})Sb$. If we take the value of the quasielastic linewidth at 5 K as its

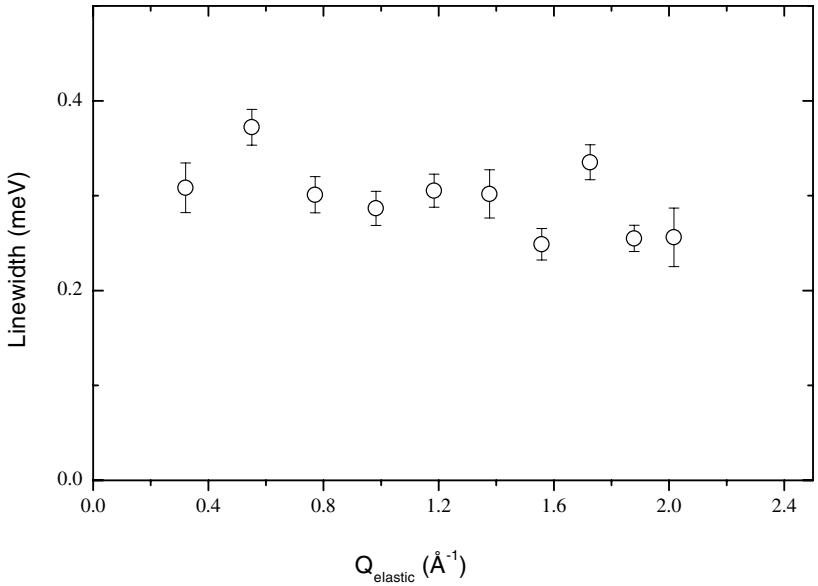

Figure 6. The Q-dependence of the quasielastic linewidth, Γ, for the data taken at 5 K (see the text).

value for $T \to 0$, then we can estimate the value of Kondo temperature, $T_K = 3.6$ K, for $Ce(Rh_{0.8}Pd_{0.2})Sb$. We acknowledge that the q-independence of the quasielastic scattering is somewhat unexpected if the NFL feature is believed to be due to a $T = 0$ quantum phase transition near an antiferromagnetic region. This thus puts $Ce(Rh_{0.8}Pd_{0.2})Sb$ in sharp contrast with $Ce(Cu_{6-x}Au_x)$, which shows a strong q-dependent feature [6]. On the other hand, we know of a few examples of antiferromagnets with weak q-dependent scattering above T_N, for example CePdSn [20].

As regards the origin of the NFL behaviour, it is very unlikely that the two-channel Kondo model is relevant to $Ce(Rh_{0.8}Pd_{0.2})Sb$ since most orbital channels of conduction electrons are non-degenerate in this case. Moreover, our observation of the SO transition indicates that any coupling between f^1 and f^2, which is the most important parameter in the two-channel Kondo model, is likely to be very weak. In fact, there is no known example of a Ce compound satisfying the criteria of the two-channel Kondo model. Concerning the Kondo disorder model, we note that our data on $Ce(Rh_{0.9}Pd_{0.1})Sb$ do not show NFL behaviour[7]. If the NFL behaviour in $Ce(Rh_{0.8}Pd_{0.2})Sb$ were to be due to Kondo disorder, we would expect the NFL behaviour to be observed over a rather wide range of concentrations. Note that a similar NFL behaviour was found for both $x = 1$ and 1.5 in $U(Cu_{5-x}Pd_x)$ [5]. However, we cannot entirely rule out the Kondo disorder model on the basis of our current data alone, as probably a very narrow range of concentration in Ce(Rh, Pd)Sb might be consistent with the Kondo disorder model. Another viable scenario is the $T = 0$ quantum phase transition model. This conclusion may be consistent with our observation of a narrow critical region with NFL behaviour. That we have observed the recovery of the usual localized behaviour for $T > 200$ K and $\hbar\omega > 15$ meV also seems to support our conjecture. Interestingly, our recent μSR measurements on $Ce(Rh_{0.8}Pd_{0.2})Sb$ show a sharp rise in the relaxation rate λ below 1.5 K, probably consistent with a $T = 0$ quantum phase transition scenario [21].

[7] Our preliminary work on $Ce(Rh_{0.7}Pd_{0.3})Sb$ suggests that it does not show E/T scaling behaviour.

In summary, we have observed a clear E/T scaling behaviour, i.e. an NFL behaviour, in the dynamic susceptibility of $Ce(Rh_{0.8}Pd_{0.2})Sb$, which is located at the critical point of a magnetic-to-non-magnetic transition. We have found that our $\chi''(\omega, T)$ can be described by a phenomenological model and the NFL behaviour is likely to arise from a $T = 0$ quantum phase transition. There is also a breakdown of the NFL behaviour in $\chi''(\omega, T)$ for $\hbar\omega \geqslant 15$ meV and $T \geqslant 200$ K.

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