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Specific heat and electronic transport properties of medium heavy-fermion compound U_2Rh_2In

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Abstract

We report on measurements of the temperature dependent specific heat, resistivity, Seebeck and Hall coefficients of U_2Rh_2In. This compound does not order magnetically down to 1.5 K and is characterized by an enhanced electronic-heat coefficient $\gamma \approx 130$ mJ/mol U K^2 at 2 K. The magnetic resistivity shows a $T^{3/2}$ law below 4 K and a broad maximum at $T^{\rho}_{\max} = 55$ K. The latter refers to the onset of coherence between U ions. The application of pressures up to 19 kbar diminishes the contribution of the spin fluctuation to the resistivity, but does not significantly change the position of the resistivity maximum. Thermopower is characterized by a deep minimum of $-30\ \mu$V K^{-1} at 10 K, followed by a sign change at 45 K. The Hall coefficient shows a maximum at $\sim T^{\rho}_{\max}$ and is dominated by skew scattering. At 2 K, the ordinary Hall coefficient $R_0 \approx 9.65 \times 10^{-10}$ m^3 C^{-1} corresponds in a one-band model to a charge carrier density of 1.33 holes/formula unit. The effective mass of the carriers at low temperatures is estimated to be $55m_e$.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

In Kondo lattice scenarios [1, 2], the transition from the nonmagnetic to the magnetic regime is often accompanied by various unusual physical phenomena such as heavy-fermion behaviour, non-Fermi liquid properties or superconductivity. Similar features were observed for a series of tetragonal compounds with the general chemical formula U_2T_2(In, Sn), where T comprises 3d, 4d and 5d transition metals. Inspecting the magnetic phase diagram of these compounds [3] one can distinguish two magnetic regimes. On the nonmagnetic side, compounds with T = Co, Ir behave as weakly temperature dependent paramagnets due to a significant strength of the exchange interaction J between the f electrons and conduction electrons. These compounds possess relatively small values of the Sommerfeld coefficient, C_p/T, less than 20 mJ/mol U K^2 [4]. On the magnetic side, U_2Ni_2(In, Sn) and U_2Pd_2(In, Sn)

order antiferromagnetically with noncollinear spin arrangements within the basal plane [5–7]. Both U_2Rh_2In and U_2Pt_2In are placed right at the magnetic to nonmagnetic border line. The latter compound exhibits a γ value of 850 mJ mol^{-1} K^{-2} and was classified as heavy-fermion compound [8]. Additionally, non-Fermi liquid characteristics were found in this material [9, 10]. In the case of U_2Rh_2In the Sommerfeld constant was deduced to be 280 mJ mol^{-1} K^{-2} [8]. Furthermore, magnetic and electrical transport measurements indicated the presence of magnetic correlations below 5 K [11]. These correlations certainly lead to a large magnetoresistance of -40% at 5 K. In order to better understand the magnetic behaviour of U_2Rh_2In, we have measured the temperature dependent resistivity under pressure, specific heat, thermoelectric power and the Hall coefficient. We will show that owing to its enhanced charge-carrier mass U_2Rh_2In can be classified as a medium heavy-fermion compound.

2. Experimental details

Polycrystalline U_2Rh_2In was prepared by arc melting the elements of stoichiometric amounts of high purity (U: 99.9 mass%, Rh: 99.99 mass% and In: 99.999 mass%) constituents, in accord with the procedure previously described in [3]. The quality of the obtained sample was examined using an energy dispersive x-ray spectrometer (PV9800) and x-ray powder diffractometer (Stoe). The data showed the sample to be single phase, belonging to the tetragonal U_3Si_2-type structure. The determined lattice parameters at room temperature $a = 7.553(3)$ Å and $c = 3.605(2)$ Å are in good agreement with the literature [3, 12]. Specific heat measurements were carried out in the temperature range 2–100 K, utilizing a thermal relaxation method. Resistivity under pressure below 19 kbar was measured in the temperature range 2–290 K by means of the standard four-probe technique. Hydrostatic pressure was generated by a piston–cylinder cell using silicon oil as the pressure-transmitting medium. Thermoelectric power was studied in the temperature range 2–300 K, using a differential method. The Hall coefficient was measured in magnetic fields up to 7 T at temperatures 2–300 K.

3. Results and discussion

Figure 1 shows the temperature dependent specific heat, C_p, of U_2Rh_2In. At 2 K, C_p/T reaches 130 mJ/K^2 mol U, slightly lower than the value reported earlier (140 mJ/K^2 mol U) [8]. For $T > 8$ K, $C_p(T)$ can be accounted for by a Debye term and an electronic contribution, i.e.

$$C_p = 9Nk_B\left(\frac{T}{\Theta_D}\right)^3\int_0^{\Theta_D/T}\frac{x^4e^x}{e^x-1}dx + \gamma_{HT}T. \quad (1)$$

A least squares fit of equation (1) to the data gives the Debye temperature $\Theta_D = 179$ K and a high-temperature electronic specific heat coefficient $\gamma_{HT} = 24$ mJ/K^2 mol U. The inset of figure 1 shows low-temperature data plotted as C_p/T versus T^2. The dashed line is a fit according to $C_p/T = \gamma_{LT} + \beta T^2$, yielding for temperatures 9–15 K $\gamma_{LT} \approx 102(5)$ mJ/K^2 mol U and β of 0.786 mJ/K^4 mol U. The latter value corresponds to $\theta_D = 183$ K, in good agreement with the high-temperature estimation. It is clear from the inset of figure 1 that below 8 K C_p/T deviates from a T^2 behaviour. Furthermore, after subtracting the electronic and the lattice contributions from the total specific heat, one observes a broad maximum centred around 4.5 K, in coincidence with that previously observed by magnetization $M(T)$ and electrical resistivity $\rho(T)$ measurements [11]. Since neutron powder diffraction experiments evidence neither magnetic order nor a crystal structure distortion down to 1.5 K [13], such an observed anomaly in $M(T)$ and $C_p(T)$ may be attributed to short-range magnetic correlations. However, we may remark that the nature of this anomaly is certainly

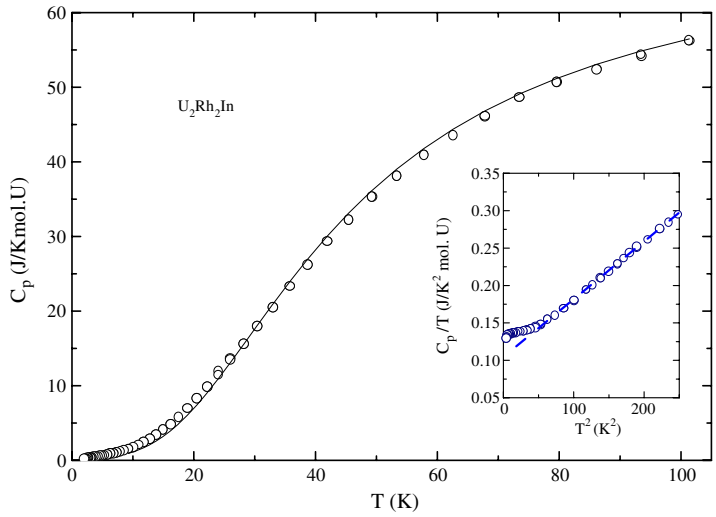

Figure 1. Temperature dependence of specific heat of U_2Rh_2In. The solid line is a fit of the data to equation (1). The inset shows the specific heat data as a plot of C_p/T versus T^2. The dashed line represents the $C_p/T = \gamma_{LT} + \beta T^2$ dependence.

different from that producing the spin-glass phenomenon, for which disorder (magnetic or atomic) is an essential ingredient [14]. A plausible interpretation would be based on the model developed by Iglesias *et al* [15] for the coexistence of intrasite Kondo exchange and intersite magnetic exchange Ruderman–Kittel–Kasuya–Yosida (RKKY) interactions with noninteger conduction-band filling. The authors have predicted a correlation temperature T_{corr} below which short-range magnetic correlations between magnetic ions occur. The model seems to explain the magnetic properties of some heavy-fermion compounds with short-range magnetic correlations, such as $CeCu_6$ and $CeRu_2Si_2$ [15].

In figure 2(a) we show the temperature dependent electrical resistivity $\rho(T)$ of U_2Rh_2In at ambient pressure. The data agree with those previously reported in [11]. In particular, $\rho(T)$ does not obey the T^2 law in the temperature range 2–4 K, and there is saturation at high temperatures. Thus, the overall temperature dependence of $\rho(T)$ of U_2Rh_2In resembles that of classical spin-fluctuation systems like UAl_2 or UPt_3 [16]. We have estimated the magnetic resistivity ρ_{mag} of U_2Rh_2In by taking the difference between the measured resistivity of U_2Rh_2In and Th_2Rh_2In. Two pronounced features of ρ_{mag} are observed: a maximum centred at about 55 K and a $T^{3/2}$ law behaviour below 4 K. A $T^{3/2}$ dependence of the resistivity has been predicted by Moriya and co-workers in terms of the self-consistent renormalization theory of spin fluctuations [17]. The authors considered the coupling of conduction electrons to antiferromagnetic spin fluctuations in the case of three-dimensional systems. For U_2Rh_2In, in addition to the contribution of spin fluctuations, short-range order effects, which are obvious from magnetic and specific heat data, have to be taken into consideration. A $T^{3/2}$ term in $\rho(T)$ is one of the characteristic features of systems showing short-range magnetic interactions [18, 19].

A valuable tool for investigating spin fluctuations is a study of the pressure dependent resistivity. Since under high pressure the contribution of the spin fluctuation to the resistivity is negligible, one can consider the contribution of the spin fluctuation to the total resistivity to be the difference of the resistivities measured under low and under very high pressures. Such a method was used by Katzman and Mydosh [20]. For U_2Rh_2In we took the difference

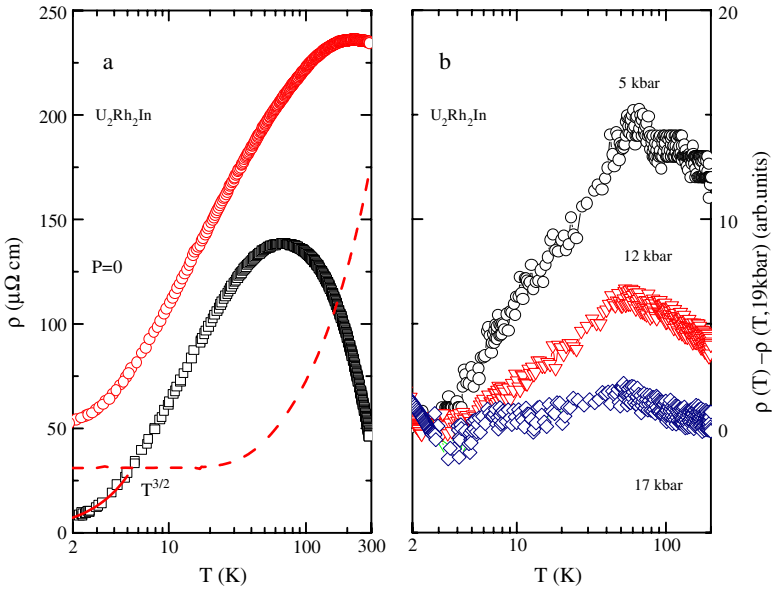

Figure 2. (a) Electrical resistivity of U_2Rh_2In (circles) and Th_2Rh_2In (dashed line) at ambient pressure as a function of temperature. The magnetic resistivity $\rho_{mag} = \rho_{U_2Rh_2In} - \rho_{Th_2Rh_2In}$ is presented as squares. The solid line is a fit to $T^{3/2}$ law. (b) Temperature dependence of the resistivity differences $\Delta\rho(T) = \rho(T, P) - \rho(T, 19\ \text{kbar})$.

$\Delta\rho(T) = \rho(T, P) - \rho(T, 19\ \text{kbar})$, assuming in addition that electron–phonon scattering is independent of applied pressure. Plotting the temperature dependence of $\Delta\rho(T)$ in figure 2(b) for several pressures we recognize that there are two temperature regions, which are distinguished by the slope $\mathrm{d}\Delta\rho(T)/\mathrm{d}T$. Below T^{ρ}_{max}, the temperature dependence of $\Delta\rho(T)$ is characterized by a positive slope while above this temperature a negative one is observed. These temperature ranges would correspond tentatively to coherent and incoherent scattering, respectively. In terms of the spin-fluctuation theory developed by Kaiser and Doniach [21], the spin-fluctuation resistivity is due to spin-flip scattering from paramagnons in a very narrow band of hybridized d- or f-electron states. This model yields $\rho \propto T^2$ at low temperatures, and at slightly higher temperatures $\rho \propto T$. The application of pressure is expected to broaden the hybridized band, leading to a decrease of spin fluctuations, but the difference $\Delta\rho(T)$ should always have a positive slope. There are several mechanisms which could account for the negative slope of $\rho(T)$. These are either related to a softening of the spin-fluctuation spectrum [22] or to a change of the 5f-electron states from a virtual bond state at high temperatures to a hybridized state at low temperatures [23]. Another mechanism is related to the Kondo effect. Kondo has shown that the spin dependent resistivity ρ_m due to the s–d exchange J_{ex}, calculated in third-order perturbation theory, is proportional to the density of states at the Fermi level $N(E_F)$ and varies logarithmically with temperature, $\rho_m \sim -J_{ex}N(E_F)\ln(1/T)$ [24]. Usually, with increasing pressure the exchange J_{ex} of Ce- or U-based compounds increases, and this leads an increase in the Kondo temperature T_K [25–27]. As a result, the resistivity maximum should shift to higher temperatures. In U_2Rh_2In the position of the resistivity maximum seems to be unaffected by the applied pressure. This observation suggests that T^{ρ}_{max} is not a function of T_K alone.

Figure 3(a) shows the temperature dependent Hall coefficient of U_2Rh_2In at 7 T. For comparison, the respective dc magnetic susceptibility χ measured with a Quantum Design

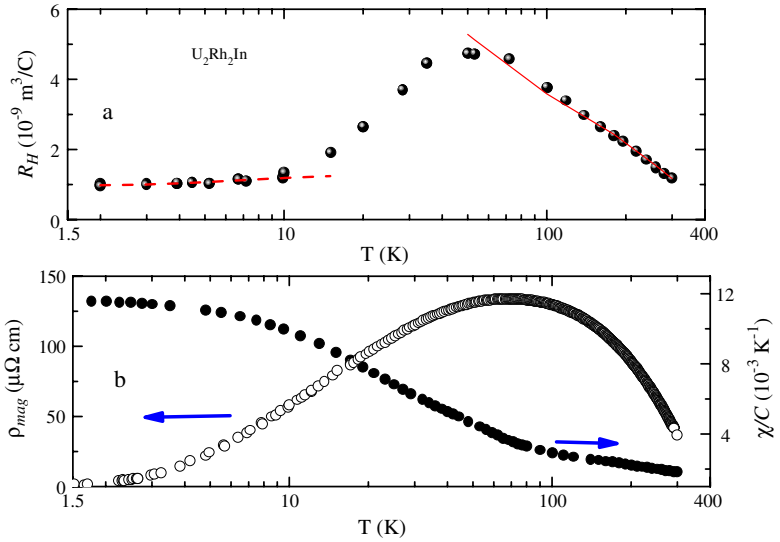

Figure 3. (a) Temperature dependence of the Hall coefficient. The solid and dashed lines are fits to equation (2). (b) Magnetic resistivity and the ratio χ/C as a function of temperature.

magnetometer at 5 T and the magnetic contribution to the resistivity are additionally shown in figure 3(b). R_H is positive at room temperature and increases with decreasing temperature, and shows a maximum at 50 K, i.e. near T^{ρ}_{max} observed in $\rho_{mag}(T)$. Clearly, the R_H behaviour of U_2Rh_2In is qualitatively the same as for other heavy-fermion compounds like $CeAl_3$, $CeRu_2Si_2$ or UPt_3, for which R_H data can be well explained by the dominant contribution of skew scattering effects, i.e. either by incoherent resonances on magnetic ions at high temperatures or by fluctuations in the coherent state at low temperatures [28]. Following Fert and Levy [28], the Hall coefficient is given by

$$R_H = R_0 + \gamma_H \tilde{\chi} \rho_{mag}, \tag{2}$$

where R_0 is the normal Hall coefficient resulting from the Lorentz motion of carriers and $R_{extra} = \gamma_H \tilde{\chi} \rho_{mag}$ is the extraordinary one originating from magnetic scattering processes of these carriers. In equation (2), the coefficient γ_H is related to phase shifts, ρ_{mag} is the magnetic resistivity and $\tilde{\chi} = \chi/C$, where C is the Curie constant and for U_2Rh_2In $C = 2.556$ cm^3 K/mol. The experimental data of the Hall coefficient can be fitted to equation (2) with $R_0 = 6.63 \times 10^{-10}$ m^3 C^{-1} and $\gamma_H = 0.766$ K T^{-1} for $T > 150$ K and $R_0 = 9.65 \times 10^{-10}$ m^3 C^{-1} and $\gamma_H = 0.038$ K T^{-1} for $T < 10$ K. The fitting parameters suggest the dominance of the incoherent skew scattering by the U 5f moments at high temperatures, while skew scattering tends to vanish at low temperatures. The results of the fits are shown in figure 3(a), as the solid and dashed lines for the high and low temperature ranges, respectively. The estimated carrier concentration based on a one-band model for U_2Rh_2In at room temperature is $n_h = 9.41 \times 10^{27}$ m^{-3}, corresponding to 1.94 holes/f.u. and a Hall mobility μ_H of 2.8 cm^2 V s^{-1}. At low temperatures, n_h and μ_H amount, respectively, to 6.47×10^{27} m^{-3} (=1.33 holes/f.u) and 19.3 cm^2 V^{-1} s^{-1}. To roughly estimate the magnitude of the effective carrier mass m^*, one can use the formula deduced from a simple free electron model: $m^* = 3\gamma\hbar^2/[(3\pi^2 n_h)^{1/3} k_B^2]$. Taking the electronic coefficient of the specific heat $\gamma_{LT} = 102$ mJ/K^2 mol U and $n_h = 6.47 \times 10^{27}$ m^{-3}, $m^* \approx 55 m_e$. Such an enhanced carrier mass suggests that U_2Rh_2In can be classified as medium heavy-fermion compound.

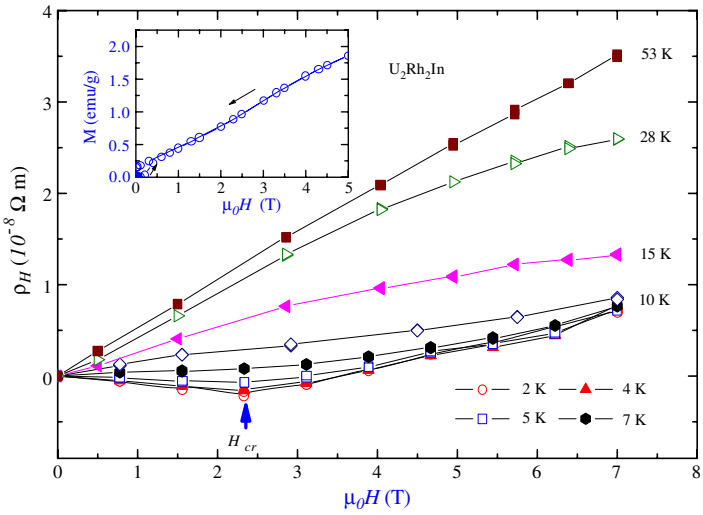

Figure 4. The dependence on magnetic field of the Hall resistivity at various temperatures. The inset shows magnetization at 1.7 K.

The Hall resistivity, ρ_{H}, as a function of magnetic field is shown in figure 4. For temperatures below 5 K, ρ_{H} shows a strong dependence on the applied magnetic field. It is negative in the low field regime but shows a profound concave dependence at higher fields; at 7 T ρ_{H} becomes positive. The change in the slope of the $\rho_{\mathrm{H}}(H)$ curves from a negative to a positive value occurs at a critical field of about $H_{\mathrm{cr}} = 2.3$ T, at the same value where the magnetization M shows a metamagnetic-like transition (see the inset of figure 4). In the temperature range 5–100 K, ρ_{H} is positive and reaches a maximum value of 3.5×10^{-8} Ω m at around 50 K.

The temperature dependent thermopower, S, of U_2Rh_2In is shown in figure 5. $S(T)$ exhibits a sharp negative minimum around 10 K and a positive knee around 75 K. Such a distinct temperature dependence of S was previously observed in various spin-fluctuating systems, for instance in Pu [29] and UAl_2 [30]. In U_2Rh_2In the S minimum of -30 μV K^{-1} occurs at $T_{\mathrm{min}} = 10$ K, compared to 15 K in UAl_2. It is interesting to note that a thermopower minimum at low temperatures has also been observed in a number of magnetically ordered f-electron compounds, such as $CeAl_2$ [30], $CeInAg_2$, $CeGe_2$ [31] and $Ce(Cu_{0.8}Au_{0.2})_6$ [32]. In all systems mentioned, the S minimum occurs at temperatures higher than magnetic phase transitions and therefore the minimum has been considered as a precursor of magnetic ordering or a signature of the presence of magnetic correlations. A theoretical description of such a minimum has been given by Fischer [33]. Furthermore, the slope $\alpha = S(T)/T$ of -2.40 μV K^{-2} observed between 2 and 4 K is also comparable to that found in UAl_2 ($\alpha = -2.13$ μV K^{-2}).

For temperatures above 125 K, $S(T)$ behaves almost linearly, with a slope of 0.023 μV K^{-2}. This is in fair agreement with the theoretical dependence (dashed line in figure 5), calculated from the charge-carrier concentration n_h. However, extrapolated experimental thermopower data for from high temperatures towards $T \rightarrow 0$ has a nonzero intercept. This fact indicates that, besides the diffusion contribution, there exist additional mechanisms, for example from magnetic interactions. In the case of UAl_2, Park and Ocko [30] interpreted the high-temperature thermopower data with help of the phenomenological resonance model used previously by Gottwick *et al* [34]. According to the latter authors the

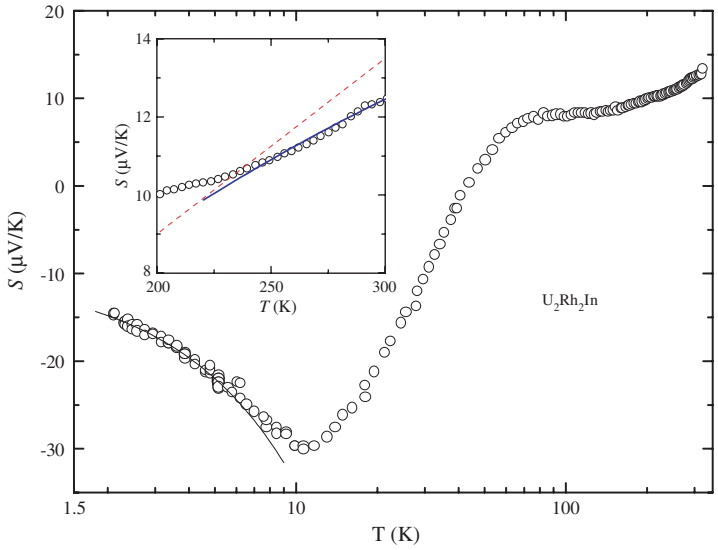

Figure 5. Temperature dependence of the thermoelectric power. The solid line with a slope of $-2.4\ \mu\mathrm{V\,K^{-1}}$ is shown. The inset shows the high-temperature S data. The dashed line is the diffusion contribution calculated from the Hall coefficient data. The solid line is the fit of the data to equation (3).

dominant contribution to S is caused by scattering between electrons of a broad s-band and a narrow f-band with the Lorentzian shape. High-temperature $S(T)$ can then be described by

$$S(T) = \frac{2(\varepsilon_f - \varepsilon_F)T/e}{\left(\frac{3[(\varepsilon_f-\varepsilon_F)^2+\Gamma^2]}{(\pi k_B)^2}\right)^2 + T^2}, \tag{3}$$

where ε_f is the position of the f-band relative to the Fermi level ε_F and Γ is the width of the resonance peak. From the data fitting we obtained $\varepsilon_f - \varepsilon_F = 9.9$ meV and $\Gamma = 97$ meV for the temperature range 230–300 K (solid line in figure 5). These values are of the same order of magnitude as those of UAl_2 (11 and 42 meV) [30].

4. Conclusions

We have presented the measurements of specific heat, resistivity, Hall effect and thermoelectric power for nonmagnetic U_2Rh_2In. The magnetic contributions to the electrical resistivity and thermoelectric power of U_2Rh_2In resemble materials having short-range magnetic interaction at low temperatures and Kondo spin fluctuations at high temperatures. The thermoelectric power, specific heat and Hall coefficient data at low temperatures display heavy-fermion characteristics, similar to those found in UAl_2 and UPt_3. The onset of coherence below 55 K and an enhanced effective mass of the carriers at low temperatures imply that U_2Rh_2In could be classified as a medium heavy-fermion compound. In our opinion, an interplay between magnetic correlations and Kondo spin fluctuations is responsible for the development of the heavy-fermion state in U_2Rh_2In.

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