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The electrodynamic response of $UCu_{3.5}Pd_{1.5}$

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Abstract. We report the results of an optical study of the $UCu_{3.5}Pd_{1.5}$ alloy over a broad frequency range from 15 to 10^5 cm^{-1}. We have evaluated the frequency dependence of the scattering relaxation rate at various temperatures and find it to increase linearly with decreasing frequency and temperature. We interpret this as a manifestation of a non-Fermi-liquid behaviour of this alloy.

1. Introduction

In relation with highly correlated electron systems, a major issue and current topic of debate is the fundamental question of whether these systems, in their normal state, may be described as simple Fermi liquids. Landau's Fermi-liquid (FL) theory is fundamental for our understanding of electron excitations in metals, as it establishes a one-to-one correspondence between the excitations of a free-electron gas and those of interacting conduction electrons in metals. One of the central points of Fermi-liquid theory is the existence of a single energy scale, the Fermi energy E_F, and for energies $E \ll E_F$ and temperatures $k_BT \ll E_F$ the electronic properties display universal behaviour. For some time it seemed that in no other solids have the predictions of Fermi-liquid theory more strikingly been realized than in the heavy-electron intermetallics [1]. In a number of these f-electron-based compounds, analyses of the low-temperature specific heat, magnetization, electrical resistivity, and dynamical susceptibility display a dependence on a single energy scale consistent with Fermi-liquid theory, although unprecedented mass enhancements of 10^2–10^3 imply strong electronic interactions [1].

More recent experimental work has indicated, however, that several heavy-electron compounds and related alloys display quite remarkable properties, which manifest much less well a 'conventional' Fermi-liquid behaviour [2]. Deviations from FL predictions have now been observed for several f-electron alloys, including $UCu_{3.5}Pd_{1.5}$ [3], $CeCu_{5.9}Au_{0.1}$ [4], $Y_{1-x}U_xPd_3$ ($x < 0.2$) [5, 6], $Th_{1-x}U_xRu_2Si_2$ ($x < 0.07$) [7] and $U_{1-x}Th_xPd_2Al_3$ [2]. Non-Fermi-liquid (NFL) behaviour is mainly deduced form the temperature dependences of the electrical resistivity, the specific heat, and the magnetic susceptibility for $T \ll T_0$: $\rho(T) = 1 - aT/T_0$ (a can be either positive or negative), $C(T)/T = (-1/T_0)\ln(bT/T_0)$, and $\chi(T) = 1 - c(T/T_0)^{1/2}$, where $|a|$, b and c are constants, and T_0 is a characteristic temperature which usually scales with the Kondo temperature T_K [2]. Such weak power laws and logarithmic divergences of the low-temperature properties of these materials suggest the existence of a zero-temperature phase transition, originating from either an unconventional moment-screening mechanism, or from a suppression of long-range magnetic or spin-glass order to $T = 0$ [2, 7–10].

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Optical investigations, extending over a broad frequency range and at various temperatures, are an efficient experimental tool for the simultaneous study of the energy and temperature dependences of intrinsic parameters characterizing these systems. Of particular relevance, in connection with the anomalous dc electrical resistivity, is the identification of the energy and temperature dependence of the transport relaxation time τ. When scattering due to phonon modes or impurities is negligible, the scattering rate $\Gamma = 1/\tau$ for an conventional FL system has the following energy (frequency ω) and temperature dependence:

$$\Gamma = 1/\tau = a(\hbar\omega)^2 + b(\pi k_B T)^2 \tag{1}$$

where a and b are frequency- and temperature-independent constants. NFL behaviour is expected to considerably affect both the temperature and frequency dependence of τ, thus leading to distinct deviations from the behaviour expressed in equation (1).

In this paper, we focus our attention on one member of this class of NFL Kondo alloys, namely $UCu_{3.5}Pd_{1.5}$. This intermetallic system crystallizes in the $AuBe_5$ structure, characterized by a periodic fcc uranium lattice with two inequivalent copper sites. The parent compound UCu_5 is a prototype Kondo-lattice antiferromagnet with a Néel temperature $T_N = 15$ K [1–3, 11]. Upon substitution of Pd for Cu ($UCu_{5-x}Pd_x$), the antiferromagnetic order is quickly suppressed, vanishing at a Pd concentration x between 0.5 and 1, and a spin-glass regime is observed for $x > 2$ [2]. From the investigation of the electrodynamic response we extract the frequency dependence of the scattering relaxation time $\tau(\omega)$ at various temperatures. We establish that at low temperatures and frequencies, $1/\tau$ decreases linearly with increasing ω and T, indicating a clear deviation from FL behaviour (equation (1)). This result is compared with our previous findings on other members of the family of Kondo alloys with conjectured NFL characteristics, like, e.g., $U_{0.2}Y_{0.8}Pd_3$ [12] and $U_{1-x}Th_xPd_2Al_3$ [13].

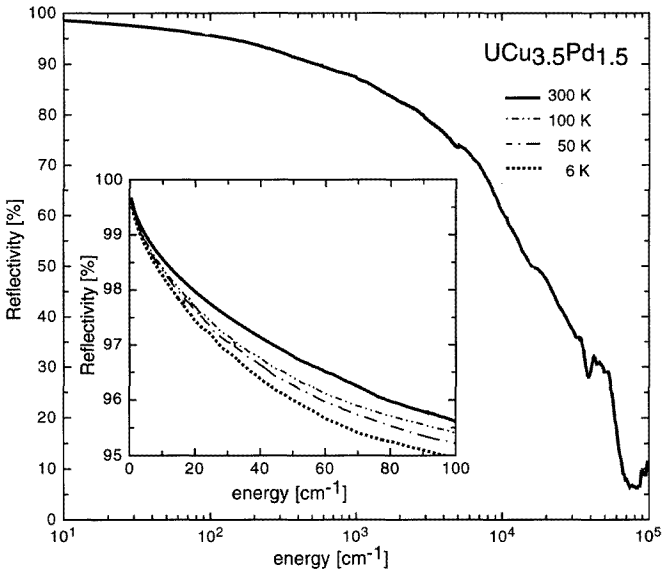

Figure 1. Reflectivity spectra of $UCu_{3.5}Pd_{1.5}$ at 300, 100, 50 and 6 K. The inset is an enlargement of the reflectivity in the FIR frequency range (linear scale).

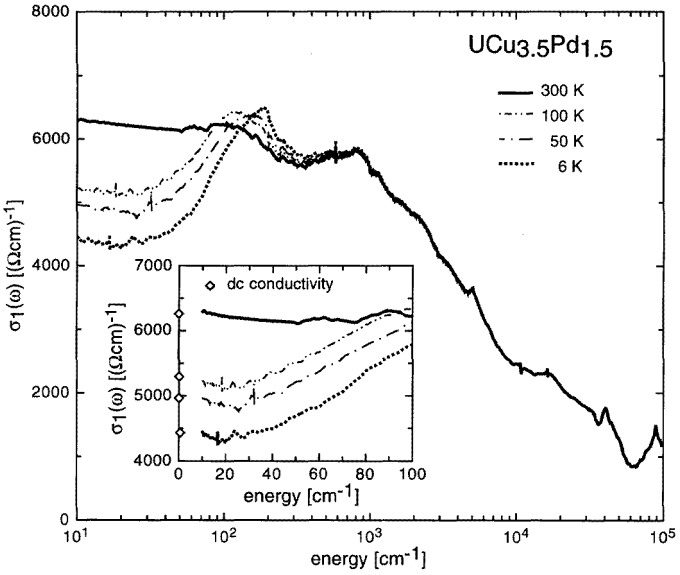

Figure 2. Optical conductivity of $UCu_{3.5}Pd_{1.5}$ at 300, 100, 50 and 6 K. The inset is an enlargement of the optical conductivity in the FIR frequency range (linear scale), showing the good agreement between the $\sigma_1(\omega \to 0)$ limit and the dc values of the conductivity.

2. Experiment and results

We measured the reflectivity $R(\omega)$ over a broad frequency range between 15 cm^{-1} to 10^5 cm^{-1}, using four spectrometers with overlapping frequency ranges [14]. The temperature dependence of $R(\omega)$ was investigated below the visible spectral range (i.e., for $\omega < 5000$ cm^{-1}). The most important frequency range for the present discussion is the far infrared (FIR), where we used a fast-scanning Fourier spectrometer, based on a Michelson interferometer, with a He-cooled Ge bolometer as the detector [14]. The optical conductivity $\sigma(\omega)$ is evaluated by the well-known Kramers–Kronig (KK) transformation of the reflectivity spectrum. Appropriate extrapolations were used above our highest-frequency limit (in the ultraviolet), while from the FIR down to zero frequency the reflectivity was extrapolated using the Hagen–Rubens law [14]. We checked that the low-frequency extrapolation does not affect the frequency dependence of $\sigma(\omega)$ in the measured spectral range (i.e., for $\omega > 15$ cm^{-1}).

Figure 1 shows the complete reflectivity spectrum at several temperatures, from which the optical conductivities, $\sigma_1(\omega)$, displayed in figure 2, were calculated. The insets of figure 1 and 2 are expanded plots of $R(\omega)$ and $\sigma_1(\omega)$ in the FIR, respectively. We note immediately that in this spectral range both $R(\omega)$ and $\sigma_1(\omega)$ decrease with decreasing temperature, while they are temperature independent at high frequencies. This is obviously in accordance with the increasing resistivity with decreasing temperature. There is also a fair agreement between the $\omega \to 0$ limit of the optical conductivity σ_1 at various temperatures and the σ_{dc}-values, obtained from the dc transport investigation (see the inset of figure 2) [2, 3, 15]. At 300 K $\sigma_1(\omega)$ is basically Drude-like up to the mid-infrared spectral range. Overlapping this typical metallic contribution is a broad maximum due to an absorption at approximately 1000 cm^{-1}. Below 100 K, apart from the suppression of $\sigma_1(\omega)$ in the

FIR, there is a displacement of spectral weight leading to a new feature at about 100–200 cm^{-1}. The high-frequency and temperature-independent excitations in the rest of the spectrum are ascribed to electronic-like interband transitions. The above characteristics and the peculiar FIR temperature dependence of $\sigma_1(\omega)$ are common to various Kondo alloys with supposed NFL behaviour [12, 13]. In fact, the results of figure 1 bear a striking similarity to those obtained for $Y_{0.8}U_{0.2}Pd_3$ [12] and more recently for $U_{1-x}Th_xPd_2Al_3$ (for $x > 0.6$) [13]. Moreover, our findings contrast with our results previously obtained on the parent compound UCu_5 [16]. In UCu_5, $\sigma_1(\omega)$ develops a narrow Drude-like resonance in the FIR with decreasing temperature, and on crossing the antiferromagnetic phase transition (i.e., $T < T_N$) there is the additional appearance of a new absorption at about 30 cm^{-1}. This latter feature was ascribed to an excitation across a gap, opening at the Fermi surface because of the antiferromagnetic transition [16].

3. Discussion

A very simple and direct interpretation of the temperature dependence of $\sigma_1(\omega)$ in the FIR might be associated with a loss of free charge carriers due to the opening of a gap. A candidate for such a gap-like feature is the new absorption developing at 100–200 cm^{-1}, which arguably might also be related to crystal-field splittings. This scenario is, nevertheless, not supported by the available experimental data [2] and neutron investigations give no evidence for distinct crystal-field excitations [2, 17].

The decrease of $\sigma_1(\omega)$ in the FIR together with the feature at 100–200 cm^{-1} are, in our opinion, different manifestations of a more general and intrinsic behaviour of these materials. Therefore, we judge it more appropriate to describe the low-frequency part of the excitation spectrum from the point of view of a generalized Drude model, where both the effective mass m^* and the scattering relaxation rate Γ are frequency dependent. This method is particularly compelling at low frequencies, where no obvious method exists for separating $\sigma_1(\omega)$ into an ordinary Drude contribution and mid-infrared modes. It will allow us to establish the temperature and frequency dependence of the scattering rates derived from either the dc conductivity or the optical response in the infrared frequency range. Hence we write [18, 19]

$$\sigma(\omega) = \frac{\omega_p^2}{4\pi} \frac{1}{\Gamma(\omega) - i\omega m^*(\omega)} \tag{2}$$

where $\omega_p = 2\pi\nu_p$ is the unscreened plasma frequency. This generalized Drude *ansatz* has already been used in several other contexts, as, e.g., for the heavy-electron compound UPt_3, revealing a Fermi-liquid behaviour [18], or even more recently for the high-T_c superconductors [20], where a 'marginal' Fermi-liquid behaviour with $1/\tau \sim \max(\omega, k_B T)$ was inferred [21, 22].

Using equation (2), we obtain the frequency dependence of $m^*(\omega)$ and $\Gamma(\omega)$ by inverting the Kramers–Kronig results for σ_1 and σ_2. Here, we limit our attention to $\tau(\omega) = 1/\Gamma(\omega)$:

$$\Gamma(\omega) = \frac{\omega_p^2}{4\pi} \frac{\sigma_1}{|\sigma_1|^2 + |\sigma_2|^2}. \tag{3}$$

Figure 3 shows $\Gamma = 1/\tau$ at various temperatures. At 300 K, $\Gamma(\omega)$ is almost constant, while at lower temperatures a quite significant frequency dependence develops. We point out that we only consider the frequency dependence of $\Gamma(\omega)$ for $\omega < 100$ cm^{-1}, i.e., well below the frequency range dominated by the absorptions at 1000 cm^{-1}.

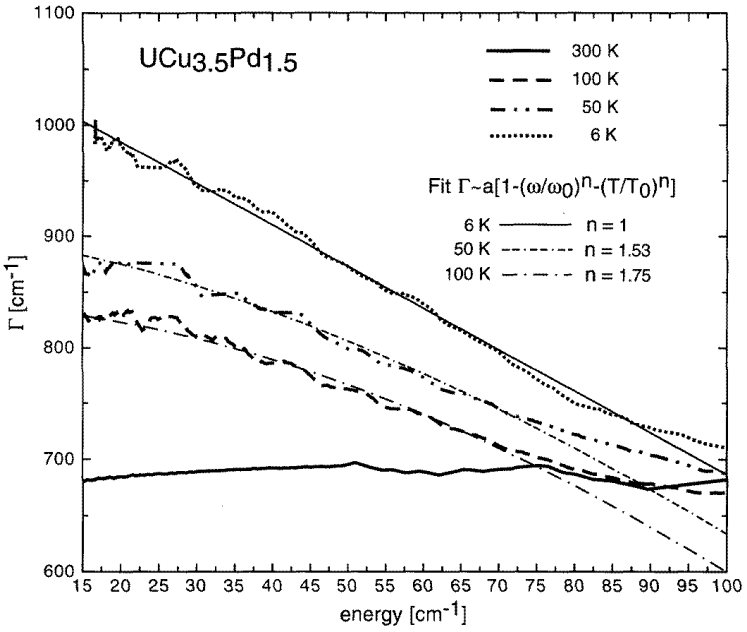

Figure 3. Frequency dependence of the transport relaxation rate Γ at various temperatures with the corresponding fits (equation (4)).

Since the temperature variation of the dc resistivity at moderately low temperatures (1 K $< T <$ 20 K) is found to vary like $1-(T/T_0)^n$, with n approximately equal to one, we are tempted to generalize this temperature dependence by including a frequency dependence for the scattering relaxation rate of the charge carriers in the form

$$\Gamma = \frac{1}{\tau} = \frac{1}{\tau_0}\left(1 - \left(\frac{T}{T_0}\right)^n - \left(\frac{\omega}{\omega_0}\right)^n\right) \tag{4}$$

where ω_0 and T_0 play the role of a cut-off frequency and temperature, respectively, and τ_0 is a constant.

At temperatures below 30 K (see, e.g., the data obtained at 6 K in figure 3) we found a good fit to $\Gamma(\omega, T)$ with $n = 1$, and values of T_0 and $\hbar\omega_0$ of about 80 K and 300 cm^{-1}, respectively. With these parameters it is possible to fit the frequency range in $\Gamma(\omega)$ extending from dc up to approximately 80 cm^{-1}. The similarity of the T- and ω-dependences implies the near equivalence of temperature and frequency or energy, establishing the temperature itself as the only relevant energy scale of the system well below the characteristic energies represented by the quantities T_0 and ω_0. Above 90 cm^{-1}, additional scattering mechanisms start to be important and the frequency dependence of $\Gamma(\omega)$ is more and more influenced by the infrared (at 100 cm^{-1}) and mid-infrared (at 1000 cm^{-1}) absorptions. This also means that the limitation of the frequency range, in which the behaviour of equation (4) is valid, implies that only a very small part of the infrared spectral weight below approximately 2000 cm^{-1} can be assigned to the carriers involved in the NFL behaviour. On the other hand, for $T >$ 30 K, the exponent n tends to increase, and the tendency of $\Gamma(\omega)$ to saturate to a constant value is clearly manifested in the FIR spectral range.

4. Conclusion

We have mapped the complete electrodynamic response of $UCu_{3.5}Pd_{1.5}$. The analysis of the data leads to frequency and temperature dependences of the scattering relaxation rate which deviate remarkably from the prediction of equation (1). Our present and previous [12, 13] work demonstrate that it is possible to monitor the different ground states of these Kondo alloys by investigating the frequency dependence of an intrinsic parameter, such as the transport relaxation rate $\Gamma(\omega)$. In the future, it would seem important to explore the frequency spectral range extending below the FIR. We expect that at such low frequencies, the low-temperature behaviour of $\Gamma(\omega)$ uncovered so far in the FIR spectral range will be further emphasized. Moreover, at these low frequencies and temperatures it should be possible to obtain additional information concerning the significance of the deviations from the power law or logarithmic divergences in $\rho(T)$ and $C(T)$ for $T < 0.3$ K [2, 5, 6, 15].

Various theoretical models have been considered to account for the microscopic origin of the NFL behaviour [2, 7–10]. As an alternative scenario to the $T = 0$ phase transition picture, it has been suggested that a two-channel Kondo effect due to electrical quadrupolar interaction might lead to a local marginal FL for which $C/T \sim \ln(\alpha T)$ and $\rho(T)/\rho(0) \sim 1 - AT^{1/2}$ [7, 8]. Specific-heat data are partially compatible with the prediction, but the linear dependence in ω and, particularly, in T of Γ does not agree with the strict theoretical expectation of the two-channel Kondo model. The distinct discrepancies in the resistive behaviour were tentatively ascribed to randomness in the system, to intersite interactions, or to a random crystal-field splitting [2]. Therefore, it remains to be seen what kind of relation may be established between our experimental optical data and the predictions of models based on the occurrence of the quadrupolar or multichannel Kondo effects, or the quantum phase transition scenario.

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References

[1] Fisk Z, Hess D W, Pethick C J, Pines D, Smith J L, Thompson J D and Willis J O 1988 *Science* **239** 33
Ott H R 1992 *Int. J. Mod. Phys.* B **6** 473
[2] Maple M B, Seaman C L, Gajewski D A, Dalichaouch Y, Barbetta V B, de Andrade M C, Mook H A, Lukefahr H G, Bernal O O and MacLaughlin D E 1994 *J. Low Temp. Phys.* **95** 225
Maple M B, de Andrade M C, Herrmann J, Dalichaouch Y, Gajewski D A, Seaman C L, Chau R, Movshovich R, Aronson M C and Osborn R 1995 *J. Low Temp. Phys.* **99** 223
[3] Andraka B and Stewart G P 1993 *Phys. Rev.* B **47** 3208
[4] von Löhneysen H, Pietrus T, Portisch G, Schlager H G, Schroeder A, Sieck M and Trappmann T 1994 *Phys. Rev. Lett.* **72** 3262
[5] Seaman C L, Maple M B, Lee B W, Ghamaty S, Torikachvili M S, Kang J-S, Liu L Z, Allen J W and Cox D L 1991 *Phys. Rev. Lett.* **67** 2882
Andraka B and Tsvelik A M 1991 *Phys. Rev. Lett.* **67** 2886
[6] Ott H R, Felder E and Bernasconi A 1993 *Physica* B **186–188** 207
[7] Cox D 1987 *Phys. Rev. Lett.* **59** 1240
[8] Sacramento P D and Schlottmann P 1989 *Phys. Lett.* **142A** 245
[9] Nozières P and Blandin A 1980 *J. Physique* **41** 193

[10] Millis A J 1993 *Phys. Rev.* B **48** 7183
Continentino M A 1993 *Phys. Rev.* B **47** 11 581
Tsvelik A M and Reizer M 1993 *Phys. Rev.* B **48** 9887
[11] Ott H R, Rudigier H, Felder E, Fisk Z and Batlogg B 1985 *Phys. Rev. Lett.* **55** 1595
[12] Degiorgi L, Ott H R and Hulliger F 1995 *Phys. Rev.* B **52** 42
[13] Degiorgi L, Wachter P, Maple M B, de Andrade M C and Herrmann J 1996 *Phys. Rev.* B **54** 6065
[14] Awasthi A M, Degiorgi L, Grüner G, Dalichaouch Y and Maple M B 1993 *Phys. Rev.* B **48** 10 692
[15] Chernikov M and Ott H R 1996 private communication
[16] Degiorgi L, Ott H R, Dressel M, Gruner G and Fisk Z 1994 *Europhys. Lett.* **26** 221
[17] Aronson M C, Osborn R, Robinson R A, Lynn J W, Chau R, Seaman C L and Maple M B 1995 *Phys. Rev. Lett.* **75** 725
[18] Sulewski P E, Sievers A J, Maple M B, Torikachvili M S, Smith J L and Fisk Z 1988 *Phys. Rev.* B **38** 5338
[19] Thomas G A, Orenstein J, Rapkine D H, Capizzi M, Millis A J, Bhatt R N, Schneemeyer L F and Waszczak J V 1988 *Phys. Rev. Lett.* **61** 1313
[20] Tanner D B and Timusk T 1992 *Physical Properties of High Temperature Superconductors III* ed D M Ginsberg (Singapore: World Scientific) p 363, and references therein
[21] Varma C M, Littlewood P B, Schmitt-Rink S, Abrahams E and Ruckenstein A E 1989 *Phys. Rev. Lett.* **63** 911
[22] Ruvalds J and Virosztek A 1991 *Phys. Rev.* B **43** 5498