Frequency and temperature dependence of the transport relaxation rate of the Kondo alloy $U_{0.2}Y_{0.8}Pd_3$: Evidence for non-Fermi-liquid behavior

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We present optical investigations of the alloy $U_{0.2}Y_{0.8}Pd_3$ over a broad frequency range from 15 to 10^5 cm^{-1}. We evaluate the frequency dependence of the transport relaxation rate at several temperatures and find it to increase linearly with decreasing frequency and temperature. We interpret this as a manifestation of a non-Fermi-liquid ground state of this alloy.

A major issue and current topic of debate in the field of highly correlated electron systems is the fundamental question whether these systems, in their normal state, may be described as simple Fermi liquids. For some heavy-electron compounds (e.g., UPt_3) this seems indeed to be the appropriate picture. Specific-heat $C(T)$ and resistivity $\rho(T)$ data supported by results of de Haas–van Alphen experiments may be well explained in this way, if strongly renormalized Fermi-liquid parameters are introduced.[1]

More recent experimental work has indicated, however, that several heavy-electron compounds and related alloys display quite remarkable properties, which may much less well be related with "conventional" Fermi-liquid behavior. Problems with Fermi-liquid-type descriptions of relevant data were met on different occasions, including materials like $UCu_{5-x}Pd_x$ for $x=1.5$,[2] $CeCu_{5.9}Au_{0.1}$,[3] and also the alloy $U_xY_{1-x}Pd_3$ for $x=0.2$.[4,5] Particularly in this latter compound, a set of data of specific-heat, magnetic susceptibility, and resistivity measurements was found to be at variance with the usual temperature dependence expected for these quantities in Fermi-liquid-type systems.[4,5] It has been suggested that a two-channel Kondo effect due to electrical quadrupolar interaction might lead to a local marginal Fermi liquid for which $C/T \sim \ln(\alpha T)$ and $\rho(T)/\rho(0) \sim 1 - AT^{1/2}$.[6,7] Specific-heat data are partially compatible with the prediction while distinct discrepancies in the resistive behavior were tentatively ascribed to randomness in the system.[8]

Little is known about the electrodynamic response of such materials. Optical investigations, extending over a broad frequency range and at various temperatures, are in principle suitable for the simultaneous study of the energy and temperature dependence of intrinsic parameters characterizing the system. Of particular relevance, in connection with the peculiar dc electrical resistivity, is the identification of the energy and temperature dependence of the transport relaxation time τ. The scattering rate $\Gamma = 1/\tau$ for a conventional Fermi-liquid system has the following energy (frequency) and temperature dependence:

$$1/\tau = a(\hbar\omega)^2 + b(\pi k_B T)^2 , \quad (1)$$

where a and b are frequency- and temperature-independent constants. Non-Fermi-liquid behavior is expected to considerably influence both the temperature and frequency dependence of τ, thus leading to distinct deviations from the behavior expressed in Eq. (1).

In this Brief Report, we present an investigation of the complete electrodynamic response of a heavy-electron-type material with conjectured non-Fermi-liquid characteristics of its electronic subsystem, for which we have chosen $U_{0.2}Y_{0.8}Pd_3$. We extract the frequency dependence of τ at several temperatures and find that $\tau(\omega)$ can be described by a frequency-dependent function similar to that describing the temperature dependence of the dc transport properties. Particularly at low temperatures and frequencies, $1/\tau$ decreases linearly with increasing ω and T, indicating that the relevant energy scale is set by the temperature alone.

The $U_{0.2}Y_{0.8}Pd_3$ specimen used for our optical investigations was prepared by arc melting in an argon atmosphere.[5] We measured the reflectivity at different temperatures and over a broad frequency range between 15 and 10^5 cm^{-1}. Four spectrometers were employed with overlapping frequency ranges. The most important frequency range for the present discussion is the far infrared (FIR), where we used a fast-scanning Fourier spectrometer, based on the Michelson interferometer, with a He-cooled Ge bolometer as detector.[9] The optical conductivity 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 FIR down to zero frequency the reflectivity was extrapolated with the Hagen-Rubens law.[9]

Figure 1(a) presents the complete reflectivity spectra at several temperatures, from which the optical conductivities $\sigma_1(\omega)$ displayed in Fig. 1(b) were calculated. We note immediately that in the FIR spectral range the reflectivity decreases with decreasing temperature, while it is temperature independent at high frequencies. Consequently, the optical conductivity $\sigma_1(\omega)$ in the FIR and its dc (i.e., $\omega \rightarrow 0$) limit drop as well.

A rather broad maximum in $\sigma_1(\omega)$ at about 1600 cm^{-1}, apparent already at 300 K, overlaps a low-frequency Drude component due to free charge carriers. Below 100 K, we note an additional broad shoulder at about 350 cm^{-1} overlapping the midinfrared absorption

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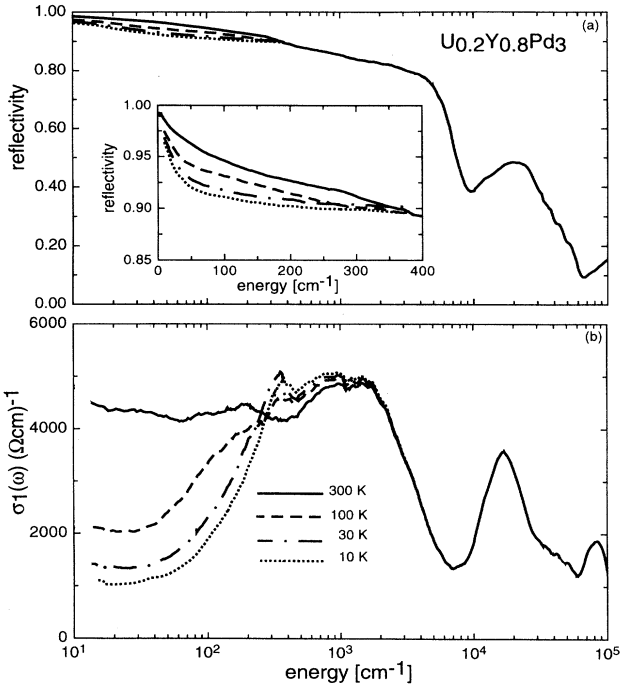

FIG. 1. (a) Reflectivity spectra at 300, 100, 30, and 10 K and (b) corresponding optical conductivity (note the logarithmic scale). The inset is an enlargement of the reflectivity in the FIR frequency range (linear scale).

mentioned above. A very simple phenomenological description of the optical conductivity may be obtained using the classical dispersion theory and associating several harmonic oscillators, i.e., Lorentzians, with the absorption maxima at 1.6×10^3, 1.8×10^4, and 8×10^4 cm^{-1}, overlapping the free-charge-carrier contribution described by the Drude model. At temperatures of 100 K and less, it was necessary to introduce an additional harmonic oscillator in order to account for the FIR shoulder at 350 cm^{-1}. This phenomenological analysis allows us to evaluate several parameters, like the unscreened plasma frequency (ν_p), the relaxation scattering rate (Γ_D) of the free charge carriers, and also the mode strength, or spectral weight, associated with the broad absorptions from the FIR up to the UV frequency range. A full report of this phenomenological analysis will be presented elsewhere.

We ascribe the high-frequency excitations to electroniclike interband transitions. For a more precise identification of these absorptions, however, we have to await a complete band-structure calculation and the corresponding joint density of states. Somewhat more puzzling is the appearance of the absorption at 350 cm^{-1} at low temperatures. It remains to be seen whether it may be related to excitations between levels of $5f$ electrons split by the crystalline electric field or simply to some phonon modes. Recent neutron-scattering measurements indicate, however, crystal-field splittings of distinctly lower energies.[8]

We now address the main issue of the present paper, namely, the possible observation of "non-Fermi-liquid" behavior by optical experiments. First, we assume that the complex optical conductivity $\sigma=\sigma_1+i\sigma_2$ whose real part is presented in Fig. 1(b) may be described by a so-called 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 midinfrared modes. It will allow us to reveal a fundamental relationship between the scattering rates derived from either dc transport properties or the optical response in the infrared frequency range. Hence we write[10]

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

where $\omega_p=2\pi\nu_p$ is the unscreened plasma frequency. For the purpose of the following discussion, we have considered $\nu_p=1.6\times10^4$ cm^{-1}, which corresponds to the plasma frequency at 300 K (i.e., the total spectral weight of the free charge carriers). It is important to recall that Eq. (2) only holds with the assumption that the enhancement of the effective mass m^* also controls the enhancement of the scattering relaxation time τ^*, i.e., $\tau^*=\tau m^*/m_b$.[11] Using Eq. (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 $\tau(\omega)=1/\Gamma(\omega)$. In Fig. 2 we present $\tau(\omega)$ for several temperatures. At 300 K, $\tau(\omega)$ is almost constant as expected, while at lower temperatures a quite significant frequency dependence develops. We point out that we consider the frequency dependence of $\tau(\omega)$ for $\omega<100$ cm^{-1}, i.e., well below the frequency range dominated by the absorptions at 350 and 1600 cm^{-1}, which we have briefly discussed above in terms of harmonic oscillators.

Since the temperature variation of the dc resistivity at low temperatures is found to follow the behavior $1-(T/T_0)^n$, with n approximately equal to 1 and a cutoff temperature $T_0=284$ K,[5] we are obviously tempted to

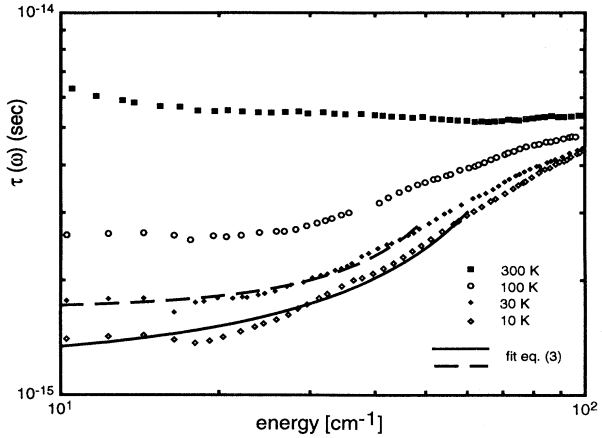

FIG. 2. Frequency dependence of the scattering relaxation time at various temperatures with the corresponding fits [Eq. (3)] at 10 and 30 K, using the parameters given in the text.

generalize this temperature dependence by including a frequency dependence for the scattering relaxation rate of the charge carriers in the form

$$\frac{1}{\tau(\omega)}=\frac{1}{\tau_0}\left[1-\left[\frac{T}{T_0}\right]^n-\left[\frac{\omega}{\omega_0}\right]^n\right], \tag{3}$$

where ω_0 plays the role of a cutoff frequency and τ_0 is a constant.

At temperatures below 20 K (see, e.g., the data obtained at 10 K in Figs. 1 and 2) we found a good fit to $\tau(\omega,T)$ with $\tau_0=1.15\times10^{-15}$ sec, $n=0.93$, $T_0=260$ K, and $\hbar\omega_0=106.23$ cm^{-1}. With these parameters it is possible to fit the frequency range in $\tau(\omega)$ extending from dc up to approximately 50 cm^{-1}. Above this frequency, additional scattering mechanisms start to be important and the frequency dependence of $\tau(\omega)$ is more and more influenced by the infrared and midinfrared absorptions. This also means that the limitation of the frequency range in which the behavior of Eq. (3) is valid implies that only a very small part of the infrared spectral weight of the absorptions at 350 and 1600 cm^{-1} can be assigned to those carriers. It is, nevertheless, remarkable that the values resulting for n and T_0 are almost identical to those evaluated from the previous dc resistivity experiment.[4,5] At temperatures above 20 K one can still apply the general expression of Eq. (3), with a different set of parameters, however. Between 30 and 60 K, $T_0=230$ K, $\hbar\omega_0=80$ cm^{-1}, $\tau_0=1.65\times10^{-15}$ sec, and $n=1.9$ must be considered.

We demonstrate the consistency of the analysis by plotting $\tau(T)$ at fixed frequency for $T<60$ K, as obtained in the following way. From the plots of $\tau(\omega)$ at various temperatures (Fig. 2) we evaluate its temperature dependence at well-defined frequencies. Figure 3 shows the temperature dependence of τ read at 15, 30, and 40 cm^{-1}, corresponding to temperatures of 21.6, 43, and 58 K, respectively, in comparison with the calculation using Eq. (3). We note that the temperature dependence of τ read at a fixed frequency between 10 and 20 cm^{-1} may be fitted quite well with the same set of parameters as used for the fit of $\tau(\omega)$ at 10 K. Similarly the parameters fitting $\tau(\omega)$ between 30 and 60 K mentioned above fit the temperature dependence of τ read at 30 and 40 cm^{-1} just as well. This obviously implies the equivalence of temperature and frequency or energy, establishing the temperature itself as the only relevant energy scale of the system well below the characteristic quantities T_0 and ω_0. It is important to observe that the linear dependence in ω and particularly in T of $1/\tau$ does not agree with the strict theoretical expectation of the two-channel Kondo model.[4,6] The dc resistivity, and consequently the temperature dependence of $1/\tau$, is expected to vary as $1-(T/T_0)^{1/2}$. It has been suggested that this discrepancy may be due to intersite interactions or random crystal-field splitting.[8]

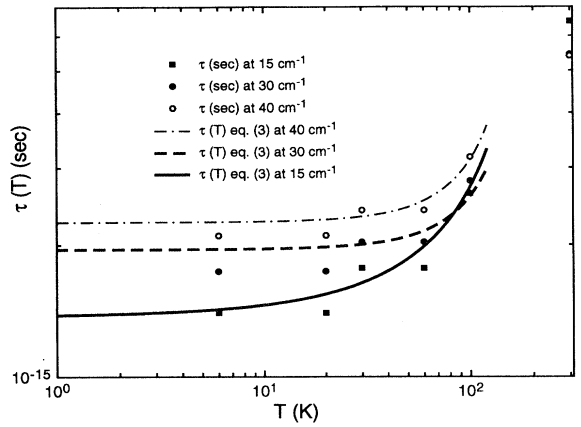

FIG. 3. Temperature dependence of the scattering relaxation time read at different frequencies (Fig. 2) and with the corresponding fits using Eq. (3), with the parameters given in the text.

We attempted to search for a manifestation of non-Fermi-liquid behavior in a suitable material mapping the complete electrodynamic response of $U_xY_{1-x}Pd_3$ with $x=0.2$. The analysis of the data leads to frequency and temperature dependencies of the transport relaxation rate which deviate remarkably from the prediction of Eq. (1), representing expectations compatible with the Fermi-liquid model. Because of lack of sufficient theoretical insight, we cannot judge whether our finding supports the interpretation of thermal and transport properties of this material based on the occurrence of the quadrupolar Kondo effect. It may well be that other possibilities for describing the electronic ground state of such systems must be considered.

In future work of this type, it seems of importance to investigate other compounds and alloys whose thermal and transport properties are indicative for unusual behavior with the aim to establish the frequency and temperature dependence of the transport scattering rate and, even more important, to address the problem of the temperature and frequency dependence of the effective mass m^*. This latter quantity is Kramers-Kronig related to $\tau(\omega)$. As indeed we find from the present study, it displays a progressive enhancement on lowering the temperature. Below 50 cm^{-1}, it starts to saturate to constant values, ranging between 20 at 100 K and 60 at 10 K. We briefly note that $m^*(\omega)$ can be fairly well approximated by the inverse of the frequency dependence of τ. A full analysis of this issue is, however, left for a future publication.

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