Relaxation-Time Enhancement in the Heavy-Fermion System $CePd_3$

W. P. Beyermann and G. Grüner
Department of Physics and Solid State Science Center, University of California at Los Angeles, Los Angeles, California 90024

and

Y. Dalichaouch and M. B. Maple
Department of Physics, University of California at San Diego, La Jolla, California 92093
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Frequency-dependent conductivity measurements are reported in the dc to 150-GHz range in the moderately heavy-fermion compound $CePd_3$. A Drude expression is found with a renormalized relaxation time τ^*, and a renormalized plasma frequency ω_p^* at low temperatures. This experiment represents the first direct measurement of these quantities, and the enhanced mass obtained from the frequency-dependent conductivity compares favorably with that determined from thermodynamic measurements.

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The various many-body effects which arise as a consequence of electron-electron interactions have been explored in detail in so-called "heavy fermion" materials.[1] The concept of an enhanced effective mass m^* is used in general to describe correlation-narrowed bands, and m^* is derived from thermodynamic parameters which sample an energy range of width k_BT around the Fermi energy at temperature T. In this communication we report experiments which focus on another parameter, the relaxation time τ^* which appears in the low-energy transport properties. In particular, we address the question of whether a Drude-type description with a renormalized effective mass and a renormalized relaxation time τ^* is appropriate for low-temperature transport that is dominated by electrons in the vicinity of the Fermi energy.

It has been recently conjectured[2-6] that the dc conductivity is not sensitive to renormalization, and

$$\sigma_{dc}=\frac{ne^2\tau^*}{m^*}\approx\frac{ne^2\tau}{m_B}, \tag{1}$$

with τ and m_B the unrenormalized relaxation time and the band mass, but no direct experiment detecting this relaxation-time enhancement has been reported. In this paper we report the first direct observation of such an enhancement, and furthermore, our experimental results are compared with optical experiments at high energies, which are not influenced by renormalization. Such a comparison leads to the evaluation of an enhanced mass, based on the frequency-dependent response alone.

To study the possible renormalization effects of the relaxation time, we have chosen the moderately heavy-fermion system, $CePd_3$. The thermodynamic effective mass is approximately[7] $40m_e$, where m_e is the free-electron mass, and both the susceptibility $\chi(T)$ and the specific heat $\gamma(T)$ indicate that the effective Fermi temperature T_{eff} is of the order of 100 K. Although various energy scales have been suggested[8] to be important in this material, they all are at least 1 order of magnitude larger than the upper measurement frequency employed by us. Consequently, finite-bandwidth effects are not expected to interfere in the frequency-dependent response which is expected to be determined by relaxation effects. In the case of a Drude description, the low-frequency conductivity $\sigma(\omega,T)$ is given by

$$\sigma(\omega,T)=\frac{ne^2\tau^*(T)}{m^*}\frac{1}{1+[\omega\tau^*(T)]^2}, \tag{2}$$

where n is the number of conduction electrons. As $\omega\rightarrow 0$, the trivial dc limit given by Eq. (1) is recovered.

The samples were prepared by the combination of stoichiometric amounts of the elements with use of a conventional arc furnace. After the samples were wrapped in Ta foil and sealed in quartz tubes filled with Ar at a pressure of 150 Torr, the specimens were annealed for 140 h at 800 °C. A spark cutter was used to excise a flat face on the samples which were then polished. X-ray analysis shows that the materials were single phase. Standard four-probe methods were employed to measure the dc conductivity on thin sections cut from the original samples. Both the x-ray analysis and the dc measurements are in good agreement with previously published results.

We performed conductivity measurements from room temperature down to 1.3 K at three selected frequencies: 35, 102, and 148 GHz.

For each frequency, a cylindrical copper cavity was constructed that resonates in the TE_{011} mode. The polished samples were clamped to the bottom of the cavity, and the resonance, which is determined by the Q of the cavity, was recorded with a signal averager by modulation of the frequency. The TE_{011} mode was selected because it has a high Q and is relatively insensitive to poor contact between the sample and the cavity body.

It is possible to calculate the Q and relate it to the surface impedances R_i of the various conductors that make

up the cavity. The expression is[9]

$$1/Q=\sum_i \gamma_i R_i, \tag{3}$$

where the γ_i are a set of geometrical constants that depend on the particular mode used; the sum contains a term for each conducting surface in the cavity. Any other additional losses in the cavity such as radiation from the coupling holes must also be added onto Eq. (3). With the sample attached, the cavity was placed in a cryostat. The resonance was measured as a function of temperature. A copper end plate was then substituted for the sample, and the Q was remeasured at various temperatures. By taking the difference in the reciprocal Q's from the two runs, we are able to eliminate any contributions to Eq. (3) which are not due to the sample.

Finally it is necessary to extract the resistivity from the measured surface impedance. Since the materials are not excellent conductors (i.e., they are not in the anomalous skin-depth regime), the surface impedance is proportional to the square root of the resistivity. The expression used to calculate the resistivity at the measurement frequency ω is[6]

$$\rho=(2/\omega\mu_0)[R_{\mathrm{Cu}}+\gamma^{-1}\Delta(Q^{-1})]^2. \tag{4}$$

$\Delta(Q^{-1})$ is the difference in $1/Q$ measured from the two runs; γ is the constant in Eq. (3) that pertains to the end plate, μ_0 the permeability of free space. R_{Cu} is the surface impedance of copper, and care must be exercised in the calculation of this quantity because at low temperatures anomalous skin-depth effects are present in copper.

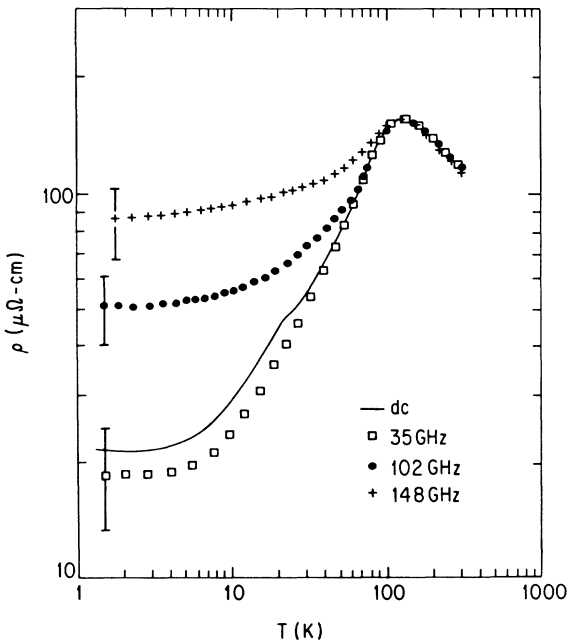

FIG. 1. Temperature dependence of the resistivity of $CePd_3$ at various frequencies. All the high-frequency data are normalized to the dc resistivity at $T=100$ K.

The conductivity measured at the three frequencies, together with the dc conductivity, is shown in Fig. 1. With data normalized to the room-temperature value, no frequency dependence is observed down to approximately 100 K, below which the conductivity is strongly frequency dependent.

With use of the experimental data displayed in Fig. 1, it is possible to evaluate the renormalized relaxation time τ^*. After rearranging Eq. (2), we have

$$[\rho(\omega)-\rho_{\mathrm{dc}}]/\rho_{\mathrm{dc}}=(\omega\tau^*)^2. \tag{5}$$

In Fig. 2, $[\rho(\omega)-\rho_{\mathrm{dc}}]/\rho_{\mathrm{dc}}$ is plotted versus ω^2 at various representative temperatures. We first note that within experimental error $[\rho(\omega)-\rho_{\mathrm{dc}}]/\rho_{\mathrm{dc}}$ is proportional to the frequency squared, and this implies that the Drude description is appropriate in the frequency range investigated by us. Figure 2 can also be used to evaluate the temperature dependence of the relaxation time. This is shown in Fig. 3, where instead of displaying τ^* as a function of temperature, we plotted $1/\tau^*$ versus the dc resistivity ρ_{dc} (both measured at the same temperature) for several different temperatures between 2.0 and 60 K. By plotting the values of $1/\tau^*$ found from the slopes in Fig. 2 as a function of the dc resistivity, we see that $\tau^*(T)$ closely follows the temperature-dependent dc conductivity up to about $T=70$ K. Above this temperature no reliable analysis can be performed because of the progressively weaker temperature dependence. The fact that $1/\tau^*$ is proportional to ρ_{dc} strongly indicates that n/m^* is independent of the temperature up to $T\simeq 60$ K; see Eq. (1). This is in agreement with observations[8] of the characteristic energy scales in this system: Both the so-called single-ion fluctuation time and the energy scale over which coherence develops among spin fluctuations

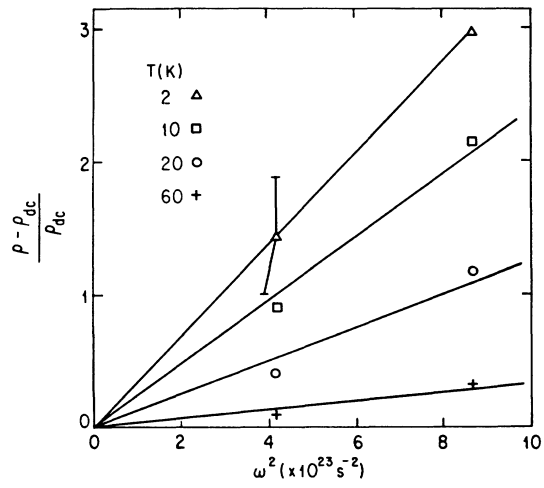

FIG. 2. Frequency dependence of the resistivity $[\rho(\omega)-\rho_{\mathrm{dc}}]/\rho_{\mathrm{dc}}$ at various temperatures. The solid lines are fits by the Drude expression, Eq. (2).

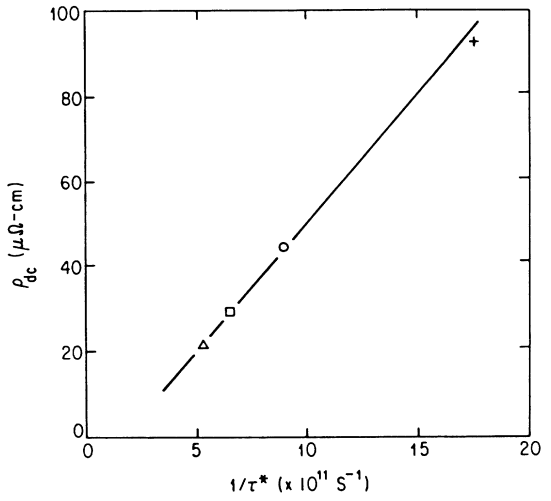

FIG. 3. (Relaxation time)$^{-1}$ as a function of the dc resistivity at various temperatures. The solid line indicates that n/m^* is independent of temperature.

on different sites exceed 40 K, and consequently both n and m^* are expected to be temperature independent at low temperatures.

We can combine the measured dc conductivity and relaxation time to define a renormalized plasma frequency

$$\omega_p^* = (4\pi\sigma_{dc}/\tau^*)^{1/2}. \quad (6)$$

At $T=2.0$ K, $\sigma_{dc}=42\times10^{15}$ sec^{-1}, and $1/\tau^*=5.4\times10^{11}$ sec^{-1}. This leads to $\omega_p^*=5.3\times10^{14}$ sec^{-1}. Renormalized effects are expected to be confined to an energy region of width ϵ_f centered around the Fermi energy,[1,5,6] and unrenormalized response occurs at frequencies $\omega > \epsilon_f/\hbar$. The characteristic energy ϵ_f is the renormalized bandwidth and is of the order of 10 meV or approximately 100 K. Consequently, the optical properties measured in the infrared spectral range, and above, reflect the unrenormalized band parameters like the plasma frequency,[10] $\omega_p=(4\pi ne^2/m_B)^{1/2}$. This has been measured by Webb, Sievers, and Mihalisin[11] employing far-infrared spectroscopy; they obtain a value of $\omega_p=3.5\times10^{15}$ sec^{-1}. The ratio,

$$\omega_p^*/\omega_p = (m_B/m^*)^{1/2}, \quad (7)$$

then yields an effective mass $m^*=44m_B$. Here we have assumed that ω_p is independent of the temperature, and this has been confirmed experimentally.[11] With this value, the carrier density can be evaluated and is found to be

$$n = m^*\sigma_{dc}/e^2\tau^* = 3.9\times10^{21}\ \text{cm}^{-3}. \quad (8)$$

The effective mass, which is obtained by the use of only the frequency-dependent conductivity data, compares favorably with m^* obtained from specific-heat and magnetic-susceptibility measurements. We also note that the evaluation of m^* from thermodynamic quantities, such as the specific heat, requires an assumption concerning the effective number of carriers since $\gamma \sim m^*(n)^{1/3}$. No such uncertainties occur in our analysis except that ω_p measures a quantity related to the average band mass.

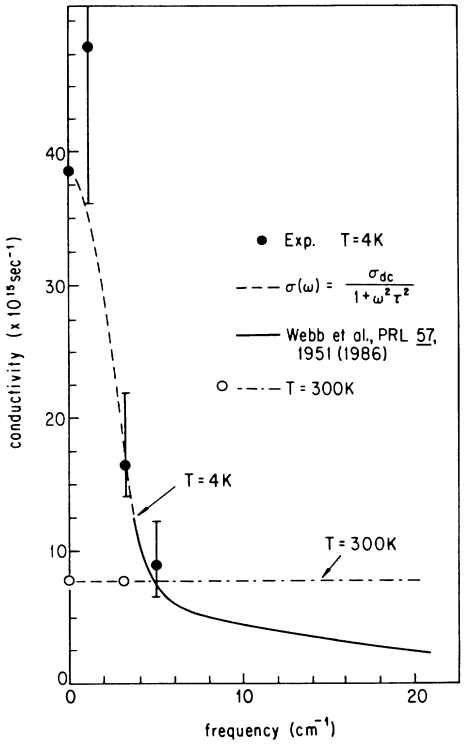

FIG. 4. Frequency-dependent conductivity at $T=4.2$ and 300 K. The solid line is from Ref. 11.

Our results are complementary to the recent far-infrared measurements of Webb, Sievers, and Mihalisin[11]; their lowest frequency corresponds to approximately 130 GHz, somewhat below our upper limit of 148 GHz. The two sets of experimental data are compared in Fig. 4 at 4.2 and 300 K. The agreement at frequencies around 4 cm^{-1} is excellent, both at low and high temperatures. The narrow resonance centered at zero frequency is clearly evident in the low-temperature region, and, as discussed before, its width is determined by lifetime effects. In contrast, only a frequency-independent conductivity is recovered at high temperatures where the various renormalization effects are removed by thermal fluctuations.

Although the enhancement of the relaxation time is well documented by our experiments, and the theoretical concepts apparently work for moderately heavy-fermion materials like $CePd_3$, several unresolved questions remain. We have assumed that the number of electrons which enter into the renormalized plasma frequency is the same as that related to the unrenormalized response. This, however, may not be *a priori* true.[10,12] The rather low carrier concentration obtained by examination of the

ω-dependent response above is a possible consequence of this uncertainty, and notice that the assumption of one f electron for each Ce atom leads to a concentration approximately a factor of 3 larger than given by Eq. (8). Also, throughout the analysis, it is presumed that the spectral width of the correlation-narrowed bandwidth is larger than our highest measurement frequency. While the weakly enhanced effective mass suggests that this is the case, small energy scales have also been suggested[13] to play an essential role in other heavy-fermion materials such as UPt_3 and U_6Fe. Additional experiments on single-crystal specimens are needed to clarify these points.

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