Low-temperature microwave response of heavy-fermion compounds

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Abstract. The electrodynamic properties of heavy fermions are distinct from those of normal metals due to the reduced transport relaxation rate that goes hand in hand with the enhanced mass. Using broadband microwave spectroscopy on thin-film samples of the heavy-fermion materials UPd$_2$Al$_3$ and UNi$_2$Al$_3$, we find that the frequency-dependent conductivity of these compounds at low temperatures follows a simple Drude prediction. The observed relaxation rates in the GHz frequency range are extremely low for a metal.

1. Introduction

Heavy-fermion materials are metals that are characterized by mobile charge carriers with an effective mass up to one thousand times the free electron mass. This mass enhancement corresponds to a reduction of the Fermi velocity, i.e. heavy fermions move much more slowly through the crystal lattice than the conduction electrons of normal metals. Consequently, the transport relaxation rate $\Gamma = 1/\tau$ of heavy fermions is reduced by orders of magnitude compared to conventional metallic electrons.[1] This relaxation rate can be observed experimentally with optical spectroscopy: in the simplest model for the optical properties of metals,[2] based on the predictions by Paul Drude, the complex optical conductivity $\sigma(\omega) = \sigma_1(\omega) + i\sigma_2(\omega) = \sigma_0/(1 - i\omega\tau)$ shows a clear frequency dependence: around the relaxation rate, the real part $\sigma_1(\omega)$ decreases from the dc value $\sigma_0$ to zero whereas the imaginary part $\sigma_2$ has a maximum (and is zero for frequencies away from $\Gamma$).

For normal metals, the relaxation rate is found at infrared frequencies, but usually a more complex frequency dependence than the simple Drude prediction is observed due to the presence of band structure, phonons, electron-electron interactions, etc.[3] Since heavy fermions are strongly interacting electron systems, their optical conductivity was also expected to show features of electronic correlations, of particular interest are signs of Fermi-liquid behavior or non-Fermi-liquid behavior.

In recent years, several heavy-fermion materials have been studied optically down to far-infrared frequencies, [4, 5, 6, 7, 8] and a characteristic suppression in $\sigma_1$, the so-called hybridization gap, was found. However, to observe the transport dynamics of the heavy charge carriers, one has to address the conductivity at even lower frequencies, in the microwave regime.
Figure 1. Literature data for cryogenic cavity experiments on CeAl$_3$ [9], CePd$_3$ [10], and UPt$_3$ ("sc" and "pc": single and polycrystals, respectively) [11]. The solid lines represent infrared data (CePd$_3$ [12]) or Kramers-Kronig analysis for combined cavity and infrared data (CeAl$_3$ [9], UPt$_3$ [11]). The data points on top of the left axis represent the low-temperature dc conductivity.

Starting already in the 1980s, a few heavy-fermion compounds have been studied with microwave cavity resonators at low temperature as shown in Fig. 1: between 10 GHz and 100 GHz there seems to be a strong decrease of $\sigma_1$, but due to the limited number of accessible frequencies (one point per cavity), a full frequency dependence could only be inferred from Kramers-Kronig arguments including infrared data. Combining thin film samples with broadband spectroscopic techniques we now have the opportunity to directly study the full frequency dependence of the microwave conductivity of heavy fermions.

2. Experiment

For the GHz frequency range, we employ a broadband microwave spectrometer in Corbino geometry.[13] So far, this approach is the only microwave experiment that allows the observation of the complex conductivity (i.e. real and imaginary parts) of heavy fermions on a broad microwave frequency range. The disadvantage of this technique is limited sensitivity: good metals can only be studied as thin films, but not as bulk crystals. Therefore, we have grown thin films of UPd$_2$Al$_3$ and UNi$_2$Al$_3$ using molecular beam epitaxy.[14, 15, 16, 17] Individual electron beam evaporators for the constituent elements are used for epitaxial deposition onto heated LaAlO$_3$(111) (in the case of UPd$_2$Al$_3$) and YAlO$_3$(112) (UNi$_2$Al$_3$) substrates. Previous experiments showed that these thin films have physical properties comparable to those of single crystals. Particularly, both compounds exhibit the known $T_c$ for superconductivity, but for the present study we concentrate on the heavy-fermion state above $T_c$.

Microwave conductivity spectra were obtained at temperatures between 1.7 K and 300 K and in the frequency range 45 MHz-20 GHz.[13] in the case of UPd$_2$Al$_3$ an additional reference measurement in the superconducting state allows reliable calibration even up to 40 GHz for temperatures below 15 K. In the case of UNi$_2$Al$_3$, the thin films grow with the c-axis of the hexagonal crystal structure in the film plane, i.e. the Corbino measurement averages different crystallographic directions. (For UPd$_2$Al$_3$ the film plane corresponds to the hexagonal plane.)

3. Results and Discussion

Two exemplary microwave conductivity spectra are shown in Fig. 2: at these low temperatures (just above the superconducting transition for UPd$_2$Al$_3$ and at the lowest accessible temperature for UNi$_2$Al$_3$), we observe a frequency-independent conductivity corresponding to the dc value for frequencies below 300 MHz, whereas $\sigma_1$ clearly decreases at higher frequencies. This roll-off can be fitted easily to the simple Drude frequency dependence.[18, 19] Here it might come as a surprise that the conductivity does not show a more complex behavior: as strongly correlated
electron systems, our compounds show very strong temperature dependences of the dc resistivity; e.g. UPd$_2$Al$_3$ at temperatures below 4 K exhibits a $T^2$-behavior that can be interpreted as a signature of a Fermi liquid, as commonly encountered in heavy-fermion materials. For Fermi liquids, a corresponding frequency dependence of the relaxation rate is expected, but we do not see any signs of this in our data. This can be explained with the extremely low relaxation rate and thus the very low frequency range that is relevant here: any frequency-dependent Fermi-liquid contribution is covered by two stronger, frequency-independent ones, namely the temperature-dependent part of the Fermi-liquid relaxation rate (the $T^2$-behavior) and the frequency- and temperature-independent scattering due to defects (the residual resistivity).

The clear presence of the Drude roll-off in our experiments unambiguously reveals the extremely low transport relaxation rate, corresponding to the reduced Fermi velocity: the charge carriers that govern the microwave response are much slower than conventional band electrons of simple metals.[18] If we now compare these conductivity spectra with the data of Fig. 1, then for UPd$_2$Al$_3$ and UNi$_2$Al$_3$ we find an even lower relaxation rate than inferred for CeAl$_3$, CePd$_3$, or UPt$_3$. This is somewhat surprising because the mass enhancement of UPd$_2$Al$_3$ and UNi$_2$Al$_3$ is only moderate compared to CeAl$_3$ and UPt$_3$. A possible explanation for this puzzle can be found by considering frequencies above 40 GHz: for UPd$_2$Al$_3$ and temperatures below 15 K, THz spectroscopy (measuring transmission through thin films) has revealed a maximum in the frequency-dependent conductivity around 120 GHz, as shown in Fig. 2. A similar feature is deduced from the cavity experiments on UPt$_3$ (green line in Fig. 1), but the current interpretation as a correlation gap closely connected to magnetism should be backed by additional studies on other heavy-fermion compounds.[20, 21]

For UPd$_2$Al$_3$, the combination of microwave and THz spectroscopy clearly shows that the Drude response of the heavy fermions takes place at frequencies below 50 GHz, but additional contributions to the optical conductivity occur already at slightly higher frequencies (around 100 GHz) - still at very low energies when compared to the optical response of normal metals.

Figure 2. Real part $\sigma_1$ of the low-temperature microwave conductivity obtained from thin films: UPd$_2$Al$_3$ (red) with Corbino spectrometer (45 MHz-40 GHz, $T=2.1$ K, sample with thickness 150 nm and $T_c=2.0$ K) as well as with THz spectrometer (above 50 GHz, $T=2.0$ K, sample with thickness 150 nm and $T_c=1.8$ K [20, 21]). UNi$_2$Al$_3$ (blue) with Corbino spectrometer (45 MHz-20 GHz, $T=1.7$ K, sample with thickness 180 nm and $T_c=1.0$ K).
Those data obtained with resonant cavities and shown in Fig. 1 thus might not be sufficient to resolve where the Drude response takes place: if also these compounds have additional optical excitations at frequencies above 10 GHz, then they might be mistaken as part of the Drude roll-off, similar to the first infrared studies on UPd$_2$Al$_3$.[22]

4. Outlook

Although our broadband experiments suggest a Drude roll-off at frequencies below 10 GHz as a general heavy-fermion feature, additional materials should be studied in this frequency range, in particular materials without magnetic order. With the advent of thin films of CeCoIn$_5$,[23] such experiments should be possible in the near future. Obviously, studies towards higher frequencies are also desirable. Furthermore, with lower accessible temperatures (the latest modifications of the Corbino spectrometer allow for continuous measurements down to 1.1 K) also the superconducting state of heavy fermions comes into reach. Finally, the anisotropy of the frequency-dependent conductivity should be studied and becomes accessible with the peculiar film growth of our UNi$_2$Al$_3$ samples: with quasi-optical THz spectroscopy, the different directions can be probed by changing the polarization of the radiation. Employing a strip geometry, anisotropy studies now also become feasible in our Corbino spectrometer.[24]

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