Influence of impurity scattering on Drude response in heavy-fermion UPd$_2$Al$_3$

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Abstract. The frequency-dependent conductivity of heavy-fermion metals can often be described within the picture of the Drude response: the transport relaxation rate is the only relevant frequency scale and, furthermore, reduced by orders of magnitude compared to normal metals. While the relaxation-time enhancement corresponds to the effective-mass enhancement in these materials, i.e. a fundamental material characteristic, the absolute value of the relaxation time depends on the details of the relevant scattering processes. Here we discuss the influence of impurity scattering on the Drude response of the heavy fermions in UPd$_2$Al$_3$ by comparing different thin film samples.

1. Introduction
Heavy-fermion materials are intermetallic compounds that show characteristic properties at low temperatures: the overall behavior is metallic, but the effective mass $m^*$ is enhanced by up to a factor of 1000 compared to the free electron mass $m_0$. This mass enhancement $m^*/m_0$ is due to the strong interaction between $f$-electrons and conventional band electrons, leading to hybridized states close to the Fermi energy, thus governing the transport properties of the materials. The feature in the electrical conductivity that corresponds to the effective mass enhancement is a reduction $\Gamma^*/\Gamma_0$ of the transport relaxation rate $\Gamma^* = 1/\tau^*$, where $\Gamma_0 = 1/\tau_0$ is the relaxation rate of the non-interacting reference material and $\tau^*$ and $\tau_0$ are the corresponding relaxation times. This scaling was expected and theoretically predicted already early in the study of heavy fermions [1, 2], but the experimental verification turned out to be challenging [3].

To directly determine the transport relaxation rate, one has to observe the full frequency dependence of the Drude response [4]: the optical conductivity $\sigma(\omega) = \sigma_1(\omega) + i\sigma_2(\omega) = \sigma_0/(1 - i\omega\tau)$ has the relaxation rate $\Gamma = 1/\tau$ as the only relevant characteristic frequency, which is manifest in a roll-off in $\sigma_1(\omega)$ and a maximum in $\sigma_2(\omega)$ at $\omega = 1/\tau$. Measuring the conductivity of a metal at frequencies around the relaxation rate is demanding for several reasons [3]: for a normal metal, the relaxation rate can be found in the far-infrared frequency range, at the low-frequency limit of conventional optical spectroscopy on metals. With the shift of the relaxation rate to even lower frequencies, the study of heavy fermions around the relaxation requires specifically optimized techniques of THz and microwave spectroscopy. Although there were early reports on Drude-like behavior with very low relaxation rate for
heavy-fermion compounds \[5, 6, 7\], the full Drude response of heavy fermions was observed only recently: for both UPd\(_2\)Al\(_3\) \[8\] and UNi\(_2\)Al\(_3\) \[9\] we could show that the low-temperature relaxation rate occurs at GHz frequencies. In the present study, we focus on the influence of impurity scattering on the Drude relaxation rate in UPd\(_2\)Al\(_3\), a well-studied heavy-fermion system with antiferromagnetic and superconducting transitions at \(T_N = 14\text{K}\) and \(T_c = 2.0\text{K}\), respectively \[10\]. Detailed studies of the optical and microwave conductivity \[8, 11, 12\] have revealed several relevant energy scales and have shown that the Drude relaxation occurs at extremely low frequencies, in the GHz range.

Following Matthiessen’s rule, the different contributions to the charge-carrier scattering rate just add. For our case, UPd\(_2\)Al\(_3\) at low temperatures, the scattering mechanisms to be considered, are impurity scattering and spin-flip scattering at the magnetic moments. The role of spin-flip scattering is evident from the strong reduction of the resistivity below the Néel temperature, within the antiferromagnetically ordered state \[10, 13\]. If we focus on the lowest accessible temperatures (just above \(T_c\) for the superconducting samples), the resistivity of all samples discussed here is dominated by temperature-independent scattering at impurities or other imperfections of the sample: in our case of thin films, scattering at the film surface can be dominant. (For the cleanest samples and just above \(T_c\), the residual resistivity contributes 70\% to the total resistivity; for the other samples this fraction is even larger.)

2. Experiment

We have prepared UPd\(_2\)Al\(_3\) thin films by coevaporation of the constituent elements in a molecular beam epitaxy system \[14\]. Previous studies have shown that thin films of very high quality can be produced by this approach \[8, 15\]. In the present study, we deliberately compare samples with different film thickness and overall quality, evident from their residual resistivity.

We have measured the frequency-dependent microwave conductivity of these samples with a broadband microwave spectrometer in Corbino geometry \[16\]. Using appropriate calibration schemes, we have determined the conductivity in the frequency range 45MHz-20GHz at temperatures down to 1.7K. Simultaneous dc resistance measurements (in 2-point geometry) allow for an in-situ comparison of dc and microwave conductivity. When required, dc resistivity was measured independently at lower temperatures.

Samples \(b\) and \(c\) have thickness of 40nm and substrate dimensions of roughly 4.5mm\(\times\)4.5mm. Sample \(a\) is much thicker, with a thickness of 150nm, and has substrate dimensions 0.9mm\(\times\)4mm. Thus, the width of sample \(a\) is smaller than the diameter of our Corbino probe. As we have shown recently \[17\], we can use this strip-shape sample geometry to even improve the sensitivity of the microwave measurements. Aside from the substrate dimensions, sample \(a\) is very similar to the sample we studied previously \[8\]. Samples \(b\) and \(c\), on the other hand, are much thinner and their sample impedance is much larger.

![Figure 1](image)

**Figure 1.** Temperature-dependent dc resistivity for different UPd\(_2\)Al\(_3\) samples. Data between 1.7K and 300K was obtained simultaneously to the microwave measurements in the Corbino spectrometer. The additional data at lower temperatures for sample \(b\) was obtained with an independent dc measurement in a \(^3\)He cryostat. For sample \(c\), no superconducting transition is expected even for lowest temperatures.
3. Results and Discussion
In Fig. 1, we present the dc conductivity of three different UPd$_2$Al$_3$ thin film samples. All of them show the temperature dependence that is characteristic for UPd$_2$Al$_3$ [10, 13, 18]: upon cooling from 300K, the resistivity first increases slightly until a maximum around 100K is reached, followed by a decrease in resistivity within the coherent heavy-fermion state. Below $T_N = 14K$, the resistivity drops steeply, since spin-flip scattering is rapidly suppressed upon cooling into the antiferromagnetically ordered state. At even lower temperatures, a superconducting transition occurs: for the highest quality sample a, $T_c = 2.0K$ corresponds to the value obtained for poly- and single-crystalline samples [10, 13, 18]. For sample b, the superconducting state is reached below 1K, whereas for sample c, with considerably higher residual resistivity, superconductivity is not expected even for lowest temperature. This dependence of $T_c$ on residual resistivity is consistent with studies on single crystals [18].

\[ \text{Figure 2. Real part } \sigma_1 \text{ of the microwave conductivity at } 2.2K \text{ for different UPd}_2\text{Al}_3 \text{ samples. The Drude response is evident from the roll-off in } \sigma_1; \text{ while this lies well within the covered frequency range for the high-quality sample a, for samples b and c the roll-off occurs close to the upper frequency limit of our measurements.} \]

The frequency-dependent microwave conductivity $\sigma_1(\omega)$ of these three samples is shown in Fig. 2 for an exemplary low temperature of 2.2K. The conductivity of sample a resembles closely that of the previously studied sample of similarly high quality [8, 19]: the full Drude response is observed, and the roll-off in $\sigma_1(\omega)$ occurs around 4GHz. For samples b and c, Drude behavior is also observed, but not as complete as for sample a: $\sigma_1(\omega)$ is constant for low frequencies and decreases for high frequencies. The suppression of $\sigma_1(\omega)$ at high frequencies can only be observed partially: for sample b, $\sigma_1$ is suppressed to 50% at a frequency of 12.5GHz, i.e. the relaxation rate is still well within our accessible frequency range. Unfortunately, at frequencies higher than 15GHz, the conductivity data is influenced by the substrate, in particular dielectric resonances. This effect is well-known (but hard to describe quantitatively) for Corbino measurements on metallic and superconducting thin films [16, 21], and is more pronounced for high-impedance samples. For sample c, we can only observe a Drude-type suppression of $\sigma_1$.
of approximately 20%; again we are hampered by substrate resonances at frequencies above 14GHz. Clearly, the Drude relaxation rate of sample $c$ is to be located outside the frequency range accessible with the present experiment. Comparing the three different samples, we can conclude that the frequency of the Drude roll-off occurs at successively higher frequencies as the dc conductivity $\sigma_0 = \sigma(\omega \to 0)$ is reduced. This indicates that the absolute value of the dc conductivity is determined by the Drude relaxation rate, which in turn is governed by the relevant scattering. For our samples at these low temperatures, the only temperature-dependent contribution is spin-flip scattering at the local magnetic moments. These are expected to be independent of sample quality. Impurity scattering, on the other hand, clearly depends on sample quality. For thin films, surface scattering can be the dominant ‘impurity’, which explains the observed behavior: while for the highest-quality samples (with film thickness 150nm) we estimate a low-temperature mean free path of 60nm, samples $b$ and $c$ with film thickness 40nm are expected to have a considerably shorter mean free path, and correspondingly a higher relaxation rate.

4. Conclusions
We have measured the low-temperature microwave conductivity of different thin film samples of UPd$_2$Al$_3$. We observe the roll-off in $\sigma_1(\omega)$ that is characteristic for the slow Drude relaxation of heavy fermions, with its extremely low relaxation rate. The absolute value of the relaxation rate increases as the dc conductivity decreases; this directly demonstrates that for the case of low temperatures, the dc conductivity depends on the impurity scattering, which in our case of thin films, can be dominated by the film surfaces.

Observing the different characteristic energy scales that occur in the optical response of UPd$_2$Al$_3$, clearly requires high-quality samples. In particular the minimum in $\sigma_1(\omega)$ around 40GHz [12], might be masked by the Drude response if the relaxation rate is rather high due to impurity scattering.

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