Far-infrared optical conductivity of CeCu$_2$Si$_2$

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Abstract
We investigated the optical reflectivity of the heavy-fermion metal CeCu$_2$Si$_2$ in the energy range 3 meV–30 eV for temperatures between 4 and 300 K. The results for the charge dynamics indicate a behavior that is expected for the formation of a coherent heavy quasiparticle state: upon cooling the spectra of the optical conductivity indicate a narrowing of the coherent response. Below temperatures of 30 K a considerable suppression of conductivity evolves below a peak structure at 13 meV. We assign this gap-like feature to strong electron correlations due to the 4f-conduction electron hybridization.

(Some figures may appear in colour only in the online journal)

1. Introduction
The investigation of the optical properties of heavy-fermion metals with 4f-electrons is focused on the charge dynamics in the far- and mid-infrared energy regions where the electronic structure is strongly influenced by a weak hybridization of localized f-electrons with conduction electrons. The relevant energy scales involved are determined by the Kondo interaction, leading to a strongly enhanced density of states at the Fermi level and the concomitant formation of heavy quasiparticle masses. A heavy-fermion state arises upon the formation of coherence among heavy quasiparticles below a lattice coherence temperature. The latter has been shown to be characteristic for the temperature dependence of the far-infrared optical response in heavy-fermion compounds like YbRh$_2$Si$_2$ and YbIr$_2$Si$_2$ [1–3]. The signature of the Kondo interaction itself was discussed in terms of a gap-like suppression in the far-infrared optical conductivity due to a conduction band ($c$–f) electron hybridization. This suppression appears at temperatures below the Kondo temperature and is related to heavy-fermion properties determining, for instance, the thermodynamics. Furthermore, the strength of the $c$–f hybridization is reflected in a broad mid-infrared peak in the range 0.1–0.3 eV at low temperatures [4]. It was shown that a dynamical mean-field approach could capture these $c$–f hybridization features in the far-infrared region (where, depending on the complexity of the hybridization, several maxima could appear) as well as in the mid-infrared region [5, 6].

To the best of our knowledge CeCu$_2$Si$_2$ does not belong to the large number of correlated electron materials whose optical properties were reported in [7], although it is one of the most investigated heavy-fermion compounds [8]. Here we report optical investigations of CeCu$_2$Si$_2$ down to energies of 3 meV and temperatures down to 4 K. This allowed us to obtain as yet inaccessible information on the low-energy heavy-fermion optical response of CeCu$_2$Si$_2$.

2. Experimental details
We have measured the optical reflectivity $R(\omega)$ at a near-normal angle of incidence on a single crystal of CeCu$_2$Si$_2$. The crystal was well characterized in its thermodynamic, transport and magnetic properties and shows superconductivity below $T_C = 0.65$ K as well as antiferromagnetic order below $T_N = 0.6$ K (‘S/A-type’)
Figure 1. Temperature dependence of the reflectivity spectrum \(R(\omega)\). The dashed line shows Hagen–Rubens behavior according to a DC resistivity of 70 \(\mu\Omega\)cm. Inset: \(R(\omega)\) at 4 and 300 K in the complete accessible range of photon energies up to 30 eV.

crystal) [9]. Those properties below 1 K do not affect the transport properties at higher temperatures, hence the DC (zero energy) electrical resistivity \(\rho_{DC}\) shows the typical temperature dependence for all types of CeCu\(_2\)Si\(_2\) samples, namely the presence of two maxima, one at 20 K due to the onset of quasiparticle coherence and one at 115 K being due to the Kondo scattering at a quasi-quartet crystalline electric field level [9]. The direction of the crystalline \(c\)-axis (tetragonal ThCr\(_2\)Si\(_2\) structure) was pointing at an angle of 15 \(\pm\) 6\(^\circ\) out of the optically investigated crystal surface. The well polished (0.3 \(\mu\)m grain) crystal was mounted behind a circular aperture with a diameter of 5 mm.

A rapid-scan Fourier spectrometer of Michelson and Martin–Puplett type was used for photon energies \(\hbar\omega\) between of 0.01–1.5 eV and 3–30 meV, respectively, at sample temperatures between 4 and 300 K. \(R(\omega)\) was extracted from two measurements: the first one measuring the sample and the second one measuring the sample after coating it \textit{in situ} with gold. Synchrotron radiation extended the energy range from 1.2 eV up to 30 eV for \(T = 300\) K [10]. Using Kramers–Kronig relations, we calculated the optical conductivity \(\sigma(\omega)\) from \(R(\omega)\). The \(R(\omega)\) spectra were extrapolated below 3 meV with the low-energy limit of the Drude model \(R(\omega) = 1 - (2\omega\rho_{DC}/\pi)^{1/2}\) (Hagen–Rubens behavior) and above 30 eV with a free-electron approximation \(R(\omega) \propto \omega^{-4}\) [11].

3. Results and discussion

The temperature dependence of the \(R(\omega)\) spectra of CeCu\(_2\)Si\(_2\) is shown in figure 1. The continuous increase of \(R(\omega)\) toward low energies indicates the response of the conduction band. The dashed line shows a Hagen–Rubens behavior with \(\rho_{DC}^{(0)}\) = 70 \(\mu\Omega\)cm (in good agreement with the measured absolute value of \(\rho_{DC}(T)\) [9, 12]) providing a reasonably good description of the low-energy data. However, inspecting the temperature evolution of \(R(\omega)\), typical metallic behavior is seen only for \(\omega \lesssim 5\) meV and below \(T \lesssim 30\) K. This peculiarity is often reported for heavy-fermion metals and is discussed in terms of a renormalization of the coherent Drude response [7].

Figure 2(a) shows the temperature dependence of the optical conductivity \(\sigma(\omega)\) below 1 eV where the charge dynamics is characterized by a pronounced temperature dependence. Between the lowest accessible energy of 3 and 150 meV, \(\sigma(\omega)\) shows a continuous decrease with decreasing temperature. As shown in figure 2(b), this energy range is characterized by the formation of a heavy plasma mode. For heavy-fermion compounds, in the low-temperature coherent state, the real part of the complex dielectric function, \(\varepsilon_1(\omega)\), is expected to show two zero-crossings from negative to positive. The crossing at lower energy \(\omega_p\) is associated with heavy plasmons, the one at higher energy with the plasma energy of uncorrelated electrons [13]. Such an analysis of \(\varepsilon_1(\omega)\) was considered, for instance, for CeAl\(_3\) [14] or for the Ce-115 system [15]. For CeCu\(_2\)Si\(_2\) (see figure 2(b)) the heavy
plasma mode $\omega^*_{p}$ already develops at $\approx 150$ K and reaches values down to about 20 meV for 4 K. Since the effective mass $m^*$ is proportional to $1/\omega^2_{p}$, $m^*$ at 4 K is about 10 times larger than that at 150 K. The plasma energy for the uncorrelated electrons is located at $\approx 1.5$ eV.

Additional information on the heavy-fermion optical response and its relation to the Kondo interaction can be inferred from the behavior of $\sigma(\omega)$ at the lowest energies. Interestingly, for $T < 30$ K $\sigma(\omega)$ shows an additional suppression in the region around $\omega_1 = 5$ meV and a concomitant evolution of a peak structure at $\omega_2 \approx 13$ meV, i.e. a gap-like feature appears. As illustrated in the inset this feature shows a temperature dependence which we estimated by using the temperature evolution of $\sigma(\omega_1)/\sigma(\omega_2)$. The overall temperature behavior of this ratio does not depend on the exact choice of $\omega_1$ and $\omega_2$. From 300 down to 30 K $\sigma(\omega_1)/\sigma(\omega_2)$ shows a continuous increase reflecting a continuous decrease of the scattering rate of the low-energy Drude part. Below 30 K, toward lower temperatures, a strong decrease of $\sigma(\omega_1)/\sigma(\omega_2)$ demonstrates the gap opening resulting in an additional suppression of $\approx 25\%$ for $\sigma(\omega_1)$ at the lowest temperature. Note, that in this temperature regime the Kondo effect with $T_K = 10$ K should also influence the charge dynamics, suggesting a relation of this gap to the presence of 4f-electrons. Hence, we attribute the suppression of $\sigma(\omega)$ below 13 meV to the opening of a hybridization gap which arises from the weak hybridization of the 4f states with the conduction electron band states as a consequence of the Kondo interaction.

The properties of the peak structure at around 13 meV are similar to those observed in other 4f-based correlated electron systems, notably in the Kondo-semiconductors YbB$_{12}$ [16] or CeFe$_2$Al$_{10}$ [17], the intermediate valence system YbAl$_3$ [18] and in the heavy-fermion systems YbRh$_2$Si$_2$ and YbIr$_2$Si$_2$ [2]. For the skutterudite heavy-fermion system CeRu$_4$Sb$_{12}$ a peak of similar type is not reported with comparable clarity [19], whereas in certain Ce-based Kondo-semiconductors comparable structures were discussed in terms of a pseudogap opening in the heavy quasiparticle band [20]. CeCu$_2$Si$_2$ seems to be the first example of a Ce-based heavy-fermion compound where such a clear gap opening in the vicinity of the Kondo temperature is observed.

The peak and shoulder structures at around 0.2 and 0.45 eV (indicated by arrows in figure 2(a)) correspond to those typically found in a variety of Ce- and Yb-heavy-fermion compounds. It was shown that their peak energies universally scale with the c-t hybridization strength [4].

The discussed signatures of strong electron correlations can be supplemented by a consideration of the energy dependence of the spectral weight which is determined by the effective charge carrier density $N_{\text{eff}}(\omega)$:

$$N_{\text{eff}}(\omega) = \frac{2m_0}{\pi e^2} \int_0^\omega \sigma_1(\omega') d\omega'$$

with $m_0$ the free-electron mass. As shown in figure 3, $N_{\text{eff}}$ becomes temperature independent for energies larger than 1 eV. This points to a correlation effect [21] because it indicates a redistribution of spectral weight up to energies much larger than the hybridization gap energy of 13 meV which characterizes the low-temperature optical response of the heavy quasiparticles.

4. Conclusion

The optical conductivity of the heavy-fermion metal CeCu$_2$Si$_2$ shows features typical for a heavy electron optical response. At the lowest accessed temperature of $T = 4$ K and the lowest energy of 3 meV signatures of the tail of a renormalized Drude peak could be observed. This is suggested by a energy dependent suppression of the optical conductivity with decreasing temperature, indicating the effect of strong electron correlations. We identified a correlation induced hybridization gap which forms below about 13 meV and for temperatures below about 30 K. This agrees with the characteristic energy of the 4f-conduction electron interaction in CeCu$_2$Si$_2$ with a Kondo temperature of 10 K. The strongly temperature dependent gap formation in the optical conductivity leads to a transfer of spectral weight toward energies up to 1 eV.

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