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Transport anomalies in CeCu$_2$Si$_2$ (*)

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Résumé. — Les propriétés de transport (résistance spécifique ρ, force thermoélectrique S, conduction thermique k) de CeCu$_2$Si$_2$ avec un niveau 4f instable et de LaCu$_2$Si$_2$ comme combinaison de référence ont été mesurées entre 1.5 K et 300 K. Pour CeCu$_2$Si$_2$, ρ atteint 220 μΩcm (à 200 K), ce qui indique un libre parcours moyen pour les électrons de l’ordre de grandeur de la distance Ce-Ce. Conformément à cela au-dessous de 100 K, nous déduisons k ~ k$_p$ (conductivité thermique des phonons). Les résultats à basse température pour S suggèrent un comportement d’un liquide de Fermi lourd avec une température caractéristique $T^* = (30±10)$ K. Au-dessus de 20 K une dépendance distincte en température de ρ et S provient de la séparation entre les niveaux du champ cristallin du niveau $j = 5/2$ de Ce$^{3+}$ avec un niveau 4f instable.

Abstract. — The transport properties (resistivity ρ, thermopower S, thermal conductivity k) of CeCu$_2$Si$_2$ with unstable 4f shell and of LaCu$_2$Si$_2$ as reference compound were measured between 1.5 K and 300 K. For CeCu$_2$Si$_2$, ρ becomes as large as 220 μΩcm (at 200 K) indicating an electronic mean free path of the order of the Ce-Ce spacing. Correspondingly, below 100 K we infer k ~ k$_p$ (phonon conductivity). The low temperature results of S suggest heavy Fermi liquid behavior with characteristic temperature $T^* = (30±10)$ K. Above 20 K, a distinct temperature dependence of both ρ and S originates in the crystal field splitting of the $j = 5/2$ state of Ce$^{3+}$ with unstable 4f shell.

The fact that in certain Ce compounds the 4f shell is unstable (via hybridization with conduction band states) is clearly visible in anomalies of the transport properties, e.g. resistivity ρ [1] or thermoelectric power (TEP) S [2]. Here we present measurements between 1.5 K and 300 K of ρ, S and the thermal conductivity k on a polycrystalline CeCu$_2$Si$_2$ sample. This compound which crystallizes in the ThCr$_2$Si$_2$ structure has an unstable 4f shell as is inferred from both previous ρ measurements [3] and recent neutron scattering experiments [4]. We have also studied LaCu$_2$Si$_2$ as a reference compound. Experimental details will be published in a separate paper [5].

Whereas the resistivity of LaCu$_2$Si$_2$ increases monotonically from the residual resistivity $\rho_n = 23$ μΩcm to the room temperature value of 84 μΩcm, the $\rho(T)$ curve of CeCu$_2$Si$_2$ is quite abnormal (figure 1):

Firstly, we observe very large resistivities up to $\rho \approx 220$ μΩcm (at 200 K) indicating maximum electron scattering with a mean free path (mfp) of the order of the Ce-Ce spacing [6]. The two distinct resistivity peaks at 20 K and 100 K and the logarithmic decrease of $\rho$ above 150 K originate [7] in (elastic and inelastic) scattering from the unstable 4f shell of the Ce$^{3+}$ ions whose $j = 5/2$ state is split by the tetragonal crystal field into three Kramers doublets.

The fast drop of $\rho(T)$ down to 1.5 K shows that the giant resistivities at higher temperatures are mainly due to thermal excitations out of the ground state of a lattice which is atomically ordered to a reasonable extent. This is valid despite the fact that our CeCu$_2$Si$_2$ sample has a residual resistivity $\rho_n$ as large as 41 μΩcm (as determined in a He$^3$ cryostat, cf. triangle in figure 1).

Like the resistivity, the thermopower of CeCu$_2$Si$_2$ also exhibits anomalous behavior (Fig. 2) : $S(T)$

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TRANSPORT ANOMALIES IN CeCu$_2$Si$_2$ shows giant (negative and positive) peaks around 20 K and 150 K and a logarithmic decrease at higher temperatures. This $S(T)$ curve is not due to phonon drag (LaCu$_2$Si$_2$ has a TEP with $0 < S < 3 \mu$V/K at all temperatures), but must be attributed to electron diffusion which is predominated by the scattering processes from the Ce ions. Again, the high temperature ($T > 20$ K) data are in qualitative agreement with theoretical expectations concerning CF split Ce$^{3+}$ ions having an unstable 4f shell [8].

![Fig. 2. — Thermoelectric power of CeCu$_2$Si$_2$ as a function of temperature.](image)

Our low temperature data suggest that below 2 K, $S = -aT$ with a very large coefficient

$$a \geq 7.5 \mu$V$K^{-2}.$$

This simple power law hints at dominating scattering processes between heavy Fermions (quasiparticles), i.e. the conduction electrons dressed by their interaction with the Ce ions which are preferentially in the lowest lying CF level. The coefficient $a$ provides an upper bound of the characteristic temperature $T^*$ ($\leq 38$ K) which is inversely proportional to the effective mass of the quasiparticles. This estimate agrees reasonably well with two more direct measures of $T^*$, i.e. the position of the giant negative TEP peak (20 K) and the residual ($T \to 0$) half width of the quasielastic neutron line (25 K) [4].

Figure 3 shows the results of the thermal conductivity experiments on CeCu$_2$Si$_2$. Also shown is the electronic contribution $k_e$ which can be qualitatively separated from the phonon conductivity $k_p = k - k_e$ under the assumption that (i) phonon-phonon Umklapp processes dominate above 300 K and (ii) the Wiedemann-Franz law is valid for electronic transport below 5 K (for details, see Ref. [5]).

Comparison of the results for LaCu$_2$Si$_2$ and CeCu$_2$Si$_2$ shows, that for the latter compound $k_p$ is only slightly smaller, whereas $k_e$ is strongly reduced. In particular, for CeCu$_2$Si$_2$ we observe a rapid drop of $k_e(T)$ below 200 K, i.e. that temperature where — according to the electrical resistivity — the electronic mfp becomes as small as possible in a metal. Thus, the very strong scattering of the conduction electrons from the unstable 4f shells in CeCu$_2$Si$_2$ leads to the hitherto unknown observation that one can readily determine the phonon conductivity of a metal over a large temperature range (which is otherwise only possible for superconductors well below $T_c$). For instance, the maximum of $k_e(T)$ at 50 K is directly visible as a pronounced peak of the measured thermal conductivity (figure 3).

To conclude, although our experiments could be performed only on a CeCu$_2$Si$_2$ sample of low metallurgical quality (as is shown by the rather large residual resistivity) we found striking anomalies in the various properties under investigation. Our observations can be interpreted as due to intrinsic transport properties of this compound containing Ce ions with unstable 4f shells.

References