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Optical study of heavy-fermion behavior in $U(Pt_{1-x}, Pd_x)_3$

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We performed an optical study of the low-energy (down to 1 meV) and low-temperature (down to 5 K) properties of single crystals of $U(Pt_{0.95}, Pd_{0.05})_3$ and of $U(Pt_{0.9}, Pd_{0.1})_3$ and we compared the results with those previously obtained on pure UPt_3. The low-temperature spectra of $U(Pt_{0.95}, Pd_{0.05})_3$ show a low-energy (a few meV) structure, very similar to the one observed in UPt_3. On the contrary, no structures have been observed in the case of $U(Pt_{0.9}, Pd_{0.1})_3$, where the spectra exhibit normal metallic behavior. The observed structures are therefore strongly connected with the heavy-fermion properties which are still present in the antiferromagnetic phase of the sample having $x = 0.05$ and disappear as the periodicity (and the coherence) is perturbed by more extensive substitution of Pt with Pd.

Among heavy fermions UPt_3 is one of the most studied compounds because of its very peculiar thermodynamic, transport and magnetic properties [1,2]. In spite of this large effort, some non-marginal aspects of such properties remain still unresolved; for example the interrelation between heavy-fermion behavior and magnetism.

One of the most interesting and effective methods in order to understand the origin of such properties, consists in slightly changing the composition and the stoichiometry of the samples in a controlled way and observing the modifications which occur. This is the case of $U(Pt_{1-x}, Pd_x)_3$, where a small amount of platinum in UPt_3 has been substituted by palladium. It turns out that the heavy-fermion properties of UPt_3 are very sensitive to such a change and rapidly disappear when the amount of Pd rises to about 10% [1,3]. For higher concentrations of palladium a phase transition occurs from the hcp structure of UPt_3 to the dhcp structure of UPd_3 [4]. We performed an optical study of the low-energy and low-temperature properties of single crystals of $U(Pt_{0.95}, Pd_{0.05})_3$ and of $U(Pt_{0.9}, Pd_{0.1})_3$ and we compared the results with those previously obtained on a similar crystal of pure UPt_3 [5]. The aim is the investigation of the low-frequency excitations of the heavy-fermion ground state.

Our experiment measures the optical reflectivity at near-normal incidence over the whole spectral range from 1 meV to 12 eV. We used four different spectrometers and the far infrared (FIR) from 1 to 84 meV has been covered by a Fourier spectrometer equipped with a cryostat and a liquid-helium cooled germanium bolometer. The measurements have been performed at room temperature and, in the FIR, also at different temperatures down to 5 K on polished surfaces. Because of the large penetration depth of the light, surface damages, which are in the order of 1 μm or less, do not affect the results in the FIR.

All the samples have been grown in the laboratories of the University of Amsterdam, and the orientation and magnetic susceptibility have been proved to be consistent with the published data of ref. [1]. A crystal with 10% of Pd has been measured on a surface lying in the basal plane, as has been used also in the case of pure UPt_3. The available sample with 5% of Pd showed a good surface containing the *c* axis, so we used polarized light in order to measure the reflectivity along this axis and in the basal plane separately. Since no measurements with polarized light were available above 4 eV, we use ellipsometric values, taken in the energy range between 1.4 and 5 eV, in order to calibrate the results of the Kramers–Kronig transformations and to check the measured reflectivities (ellipsometry is less sensitive to the surface quality). On this sample of $U(Pt_{0.95}, Pd_{0.05})_3$, which showed antiferromagnetic order below $T_N = 5.8$ K, we performed measurements at several temperatures ranging from 4.5 to 12 K, but no significant differences have been found. The data have been analyzed in terms of Kramers–Kronig transformations, in order to calculate the optical conductivity and the dielectric functions ϵ_1 and ϵ_2. The measured reflectivities and the optical conductivities obtained at 5 K are shown, respectively in figs. 1 and 2.

The main feature of the low-temperature spectrum of UPt_3 was the existence of low-energy structures [5]. In the optical conductivity we observe a minimum at about 2 meV which separates a low energy region with free carrier behavior and the first optical structure at 4 meV. The oscillator strength of this transition and the plasma frequency of the free carrier was estimated, respectively, to $f = 0.018$ and $\omega_p = 280$ meV [5]. Such features have been associated with the formation of narrow quasiparticle bands (at least two, between which the optical transition occurs and with the Fermi level in one of them) originating from the interaction and

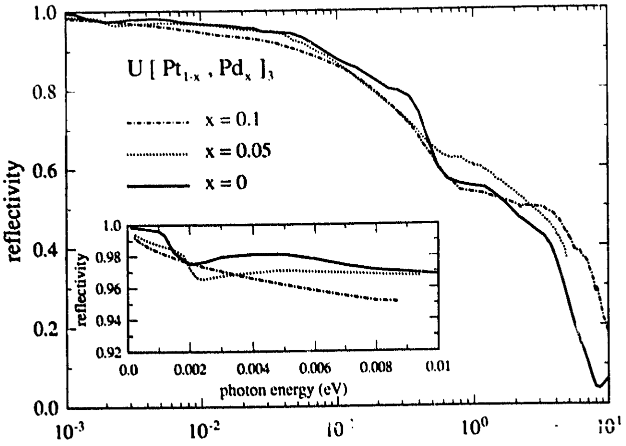

Fig. 1. Optical reflectivity of $U(Pt_{1-x}, Pd_x)_3$ at 5 K for three different stoichiometries. In the inset the FIR part of the spectra is shown.

hybridization of the f electrons of uranium with the conduction electrons. This implies heavy effective masses of the carriers. The data obtained on the sample with 5% of Pd show substantially the same characteristics. The minimum in the conductivity appears slightly shifted towards higher energies, as well as the peak of the structure which is a bit lower and broadened with respect to the one of pure UPt_3. Correspondingly the value of the dc conductivity, that is the limit at zero frequency of the optical conductivity, is considerably lowered. The existence of antiferromagnetic order does not seem to affect such a configuration. The plasma frequency determined with a fit of a Drude model has a value which is the same (or only a bit lower) than ω_p of UPt_3; therefore the decrease of the conductivity seems to mainly originate from an increase of scattering (or disorder) in the system. The low-energy feature disappears completely in the spectrum of $U(Pt_{0.9}, Pd_{0.1})_3$. In this case no significant differences can be observed between the measurements performed at 5 and 300 K respectively. At both temperatures and at the lowest energies the reflectivity is a monotonic, structureless function of the energy, consistent with the behavior expected from the Hagens–Rubens relation. As far as we know, data of the dc conductivity are available only on polycrystalline samples [3]. Our data would indicate a lower value, consistent with a conductivity in the basal plane lower than along the c axis. The first structure in the spectrum of the optical conductivity of the 10% sample is centered at about 30 meV and should correspond to a collection of structures observed in the same spectral range in UPt_3 [5]. Due to the presence of this structure at low energies, it is difficult to estimate the plasma frequency which should be in any case ranging between 1.5 and 3.5 eV. Anyway, at low temperature there is no indication of any change with respect to the normal metallic behavior at room temperature.

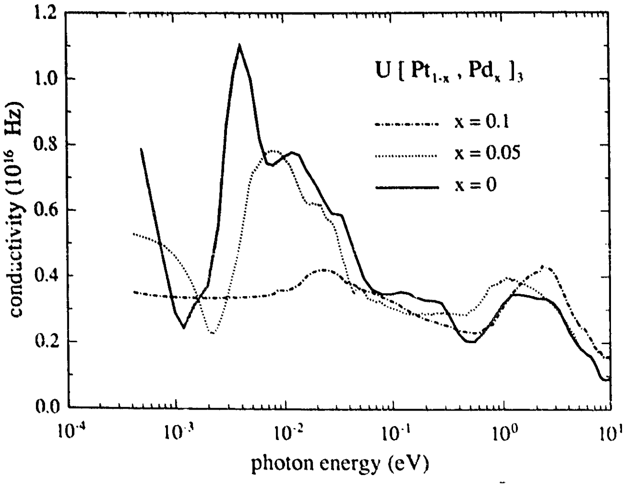

Fig. 2. Optical conductivity of $U(Pt_{1-x}, Pd_x)_3$ at 5 K for three different stoichiometries.

As a matter of fact, the substitution of Pt with Pd can alter the system in two ways. One is the direct change of the hybridization process due to the volume change. Such an effect has been claimed to be responsible of the antiferromagnetism entering the system $CeCu_{6-x}Au_x$ [6]. A second effect is the breaking of the periodicity of the hexagonal cell of UPt_3. The disorder introduced in this way destroys the coherence in the lattice of the interaction between conduction and f electrons. The f electrons of U tend to localize and quasiparticle bands are no more observable. This, in effect, is the case for UPd_3 [7].

It is interesting to note that, when the two quasibands disappear, the long-range magnetic order is lost too. We then connect these features to the loss of long-range periodicity. We could also suggest that the antiferromagnetic order for $0 < x < 7\%$ [2,3] with its low magnetic moment $(0.02\mu_B)$ is intrinsically connected with this coherent hybridization of f and d bands and thus the formation of narrow quasiparticle bands. Anyway, these bands (and the low-energy structures which appear at low temperature in the optical conductivity of UPt_3) are an intrinsic feature of the heavy-fermion state and disappear when heavy-fermion character is lost.

This is not in contrast with the observed increase of the specific heat passing from UPt_3 to $U(Pt_{0.9}, Pd_{0.1})_3$ [1]. In $U(Pt_{0.9}, Pd_{0.1})_3$ the periodicity is lost, but we still have a Kondo resonance at the Fermi level. However, in this nonperiodic arrangement the quasibands do not split, the γ value can be high, but the optical transition giving evidence of split quasibands is absent. In fact the resistivity is Kondo-like (steadily increasing for $T \to 0$) whereas in a heavy fermion the resistivity decreases to 0 for $T \to 0$.

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