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Optical Investigation of the Electrodynamic Response of UPd_2Al_3.

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Abstract. - We have investigated the electrodynamic response of UPd_2Al_3 both above and below the antiferromagnetic phase transition at $T_N = 14$ K. At low temperatures, we observe the formation of a low-frequency narrow resonance, which indicates the development of the many-body coherent state. However, we do not find any evidence of a gap absorption associated with a spin-density-wave state, which would develop if the state arised as a consequence of a Fermi surface instability.

The electrodynamics of several heavy-fermion (HF) metals has been explored in detail. In the coherent state, the optical conductivity shows a narrow resonance, indicating an enhanced relaxation time τ^*. Moreover, the small spectral weight of the resonance is (through sum rule arguments) related to a large effective mass m^*. However, significantly less is known about the electrodynamics of the superconducting and magnetic states of these materials. The prominent examples of HF superconductors, which exhibit quite different magnetic properties, are $CeCu_2Si_2$, UPt_3, UBe_{13} and URu_2Si_2. Of particular interest is URu_2Si_2, which attracted a lot of attention as the first HF metal showing both superconductivity ($T_c = 1$ K) and magnetic order ($T_N = 17.5$ K) [1]. The d.c. resistivity increases at the antiferromagnetic transition [2]—indicating the partial removal of the Fermi surface—and optical experiments give evidence for a single-particle gap [3]. Both suggest that the magnetic state develops as a consequence of a Fermi surface anomaly, similar to the one at the spin-density-wave (SDW) transition in Cr. Very recently, a systematic search led to the discovery of the coexistence of antiferromagnetism and superconductivity in UNi_2Al_3 ($T_N = 4.6$ K and $T_c = 1$ K) and UPd_2Al_3 ($T_N = 14$ K and $T_c = 2$ K) [4,5].

In this paper, we discuss our optical experiments on UPd_2Al_3. The d.c. magnetic susceptibility exhibits an anomaly (kink) at 14 K and a remarkable drop at about 2 K, where also the specific heat shows clear anomalies. The behaviour of $\rho(T)$ (see fig. 1) is more subtle;

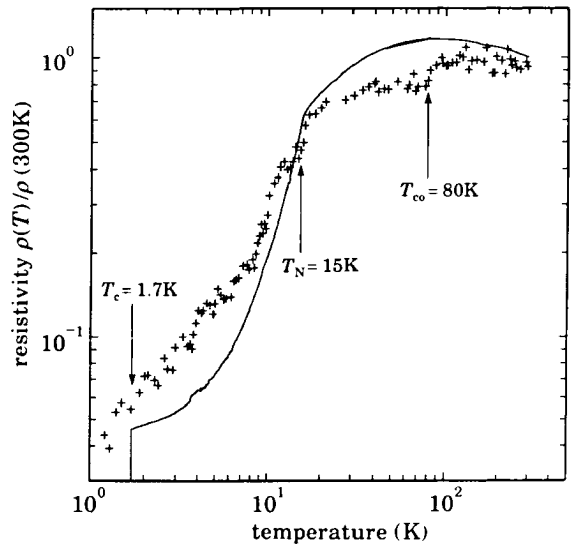

Fig. 1. – Temperature dependence of the electrical resistivity at d.c. for UPd_2Al_3 (——) and 100 GHz (+) normalised to the room temperature value.

it displays a maximum at about 80 K, ascribed to the so-called coherence temperature T_{co}, a shoulder at T_N, and the sharp drop to zero at T_c. Besides the two phase transitions at T_N and T_c, the overall behaviour of $\rho(T)$ (see fig. 1) bears a close similarity to the transport properties of other HFs. Above $T_{co} = 80$ K, $\rho(T)$ decreases with increasing temperature in a manner similar to that observed in metals containing isolated magnetic impurities, thus suggesting a Kondo scattering mechanism. Below T_{co}, where the many-body effects progressively develop, there is the antiferromagnetic (AFM) transition at 14 K, and $\rho(T)$ reflects the freezing-out of spin-disorder scattering.

The specimen of UPd_2Al_3 was prepared following the procedure described by Geibel *et al.* [4,5]. The polycrystalline samples were prepared by arc melting in an argon atmosphere and then vacuum annealed for 7 days at 900 °C, in order to avoid remaining secondary phases and improve the lattice perfection. X-ray diffraction, magnetization and resistivity measurements were performed in order to characterise our specimens. For instance, an X-ray powder diffraction measurement showed that UPd_2Al_3 crystallises in the hexagonal $PrNi_2Al_3$ structure with lattice parameters $a = 5.365$ Å and $c = 4.186$ Å [5], and the relative resistivity ρ *vs.* temperature T, displayed in fig. 1, clearly shows the main features described above. Particularly, the Néel temperature can be seen right below 15 K; below that temperature the resistivity can be fitted by the relation described in ref. [6]. Moreover, the results from the millimetre wave measurements, performed at 35 GHz and 100 GHz, are similar to the d.c. behaviour (fig. 1). Other sample characterisation gave results in accord with previous investigations [5], thus demonstrating the good quality of our samples.

The reflectivity $R(\nu)$ has been measured on a broad frequency ($\omega = 2\pi\nu$) range between 14 cm^{-1} and 10^5 cm^{-1}, using four spectrometers with overlapping frequency ranges. In the far-infrared (FIR) we made use of a Bruker IFS113v Fourier interferometer with a Hg arc light source and He-cooled Ge-bolometer detector, while from the FIR up to the mid-IR a fast scanning Bruker interferometer IFS48PC was used. In the visible spectral range a home-made spectrometer based on a Zeiss monochromator was employed, and in the

ultraviolet we used a McPherson spectrometer. From the mid-IR down to the FIR we have used the reflectivity of gold as a reference. We measured two crystals, with polished surfaces. We have observed that the polishing procedure only increases the overall reflectivity without any other qualitative changes, relatively to the measurements previously performed on the unpolished surfaces.

Figure 2*a*) displays $R(\nu)$ (note the logarithmic scale) at several significant temperatures above and below T_{co} and T_N. The inset shows the FIR frequency range in detail. We remark that $R(\nu)$ at temperatures close to and below T_{co} is generally lower than $R(\nu)$ at 300 K. Furthermore, around 100 cm^{-1} the reflectivity at 20 K merges with that at 300 K, while there is a crossover to higher reflectivity at 6 K and 15 K. This is a quite general behaviour in the optical properties of HFs (see below). However, $R(\nu)$ in UPd_2Al_3 does not show any sharp changes when the temperature decreases below T_N, and $R(\nu)$ does not display a feature that can be related to a SDW gap excitation, in contrast to what was found in URu_2Si_2 [3].

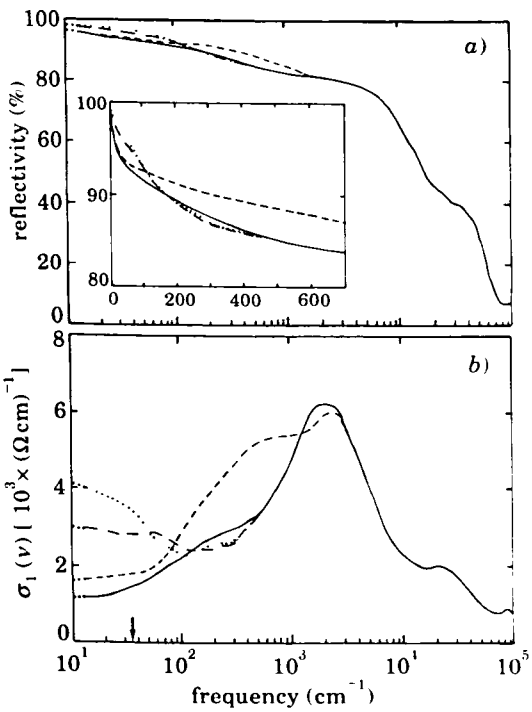

Fig. 2.

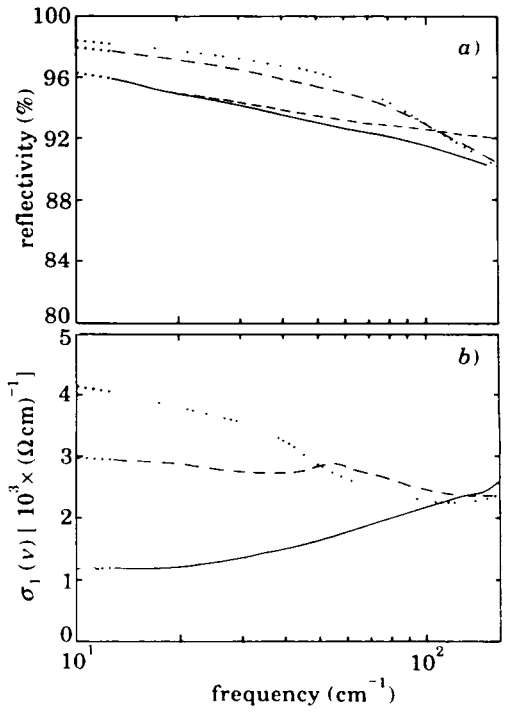

Fig. 3.

Fig. 2. – *a*) Reflectivity of UPd_2Al_3 at several temperatures (– – – 300 K, —— 20 K, –·–·– 15 K, ····· 6 K, ○ Hagen Rubens) in the whole investigated frequency range (note the logarithmic scale). The inset displays the FIR part (linear scale) of the reflectivity on an expanded scale. *b*) Optical conductivity obtained through Kramers-Kronig transformation of the spectra in *a*). The arrow marks the expected position of the SDW gap $2\Delta = 3.52 k_B T_N$.

Fig. 3. – *a*) Reflectivity and *b*) optical conductivity in the FIR (logarithmic scale), enhancing the reflectivity behaviour which leads to the narrow Drude-like resonance at low frequencies in the optical conductivity. The arrow marks the expected position of the SDW gap $2\Delta = 3.52 k_B T_N$. (Notations are the same as in fig. 2.)

The optical conductivity $\sigma_1(\nu)$ is obtained through Kramers-Kronig (KK) transformation of $R(\nu)$. The $R(\nu)$ spectra have been extrapolated to zero frequency by means of the Hagen-Rubens relation and beyond the highest measurable frequency (~ 12 eV) by the well-known $R(\nu) \sim 1/\nu^2$ and $R(\nu) \sim 1/\nu^4$ (for $\nu > 3 \cdot 10^5$ cm^{-1}) extrapolations. We remark that our $R(\nu)$ spectra are consistent with a Hagen-Rubens extrapolation making use of $\sigma_{\text{d.c.}}$ values somewhat lower than the published ones [5, 6]. This can also be seen in the d.c. limit (*i.e.* $\sigma_1(\nu \to 0)$) of our optical conductivity (see fig. 2*b*) and 3*b*)). Nevertheless, a multiplication of the $R(\nu)$ spectra by a factor of 1.01 (which corresponds to a change of about 1% in the FIR at ~ 20 cm^{-1}, roughly the limit of precision of the experiments) would remarkably improve the agreement between the d.c. limit of the optical properties and the d.c. transport data. However, since we are interested in the relative temperature dependence (*i.e.* in the behaviour above and below T_N), we do not perform here this renormalization of our raw spectra, as this will not change our main conclusion. The result of our KK transformation is shown in fig. 2*b*). At 300 K we see a frequency-independent response at low frequencies, a fairly broad mid-IR absorption at approximately 2000 cm^{-1}, and several absorptions at high frequencies (*i.e.* above 10^4 cm^{-1}). We believe that these high-frequency absorptions are electronic interband transitions; band structure calculations would be of great help for a definite identification of these excitations. The features of $\sigma_1(\nu)$ can be well fitted with a Drude term and harmonic oscillators (detailed fits will be presented elsewhere). We also notice the removal of spectral weight between approximately 100 and 800 cm^{-1} by decreasing the temperature from 300 K to 6 K. The removed spectral weight shifts to higher frequency at the mid-IR excitation, so that the *total* spectral weight is conserved (within the experimental error). Furthermore, at low temperatures, in accordance with the peculiar FIR behaviour of $R(\nu)$, we observe the formation of a temperature- dependent narrow Drude-like resonance centred at zero frequency. This aspect is underlined more clearly in fig. 3, where the $R(\nu)$ and the corresponding $\sigma_1(\nu)$ below 200 cm^{-1} and at different temperatures are displayed on an expanded scale. Later, we will return to the discussion of this important feature.

Next we discuss our experimental results. We first note the complete absence of an absorption, the onset of which would be coincident with T_N. As pointed out above, we do not find a feature at (or near to) $2\Delta = 3.52 k_B T_N$, which could be ascribed to a so-called SDW gap, as observed for example in Cr [7] or URu_2Si_2 [3]. This peculiar behaviour of $\sigma_1(\nu)$ is indicative of the fact that the antiferromagnetic transition is *not* the consequence of the $2k_F$ nesting of the Fermi surface (or part of it) and thus is not an instability leading to a SDW ground state. The d.c. resistivity also indicates the absence of a Fermi surface anomaly, and $\rho(T)$ reflects the freezing-out of the spin-flip scattering mechanism and the increase of the relaxation time τ [1].

The temperature dependence in $\sigma_1(\nu)$ is due to a temperature-dependent narrow Drude-like resonance in the FIR range (fig. 2*b*) and 3), ascribed to the optical conductivity of the heavy quasi-particle. This behaviour is reminiscent of what has been found in other HF systems, as, *e.g.*, in UPt_3 [9, 10] and $CeAl_3$ [11, 12]. The narrow resonance centred at $\omega = 0$

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[1] The reader should note that the SDW single-particle gap (which is missing in $\sigma_1(\nu)$ of UPd_2Al_3) should not be confused with the gap of about 80 K (or 40 K in ref. [6]) evaluated from the analysis of the $\rho(T)$ measurement below T_N. The latter corresponds to the gap in the magnon dispersion relation and has nothing to do with the gap which should open at the Fermi surface as a consequence of a SDW instability. See, *e.g.*, ref. [6] and [8].

can be fitted with the so-called renormalized Drude expression [11, 12]

$$\sigma(\omega) = \frac{ne^2\tau^*}{m^*}\frac{1}{1-i\omega\tau^*}, \tag{1}$$

where m^* and τ^* (assumed in the present analysis to be frequency independent) are the effective mass of the heavy quasi-particles and the renormalized relaxation scattering time, respectively. At 6 K, we find good agreement between experiment and the fit with $h\nu_p^* = 0.47$ eV and $\Gamma_D^*(\sim 1/\tau^*) = 7.1$ meV, while at 15 K $h\nu_p^* = 0.754$ eV and $\Gamma_D^* = 0.025$ eV. It is interesting to observe that $h\nu_p^*$ can be also calculated from the Pippard formula for the penetration depth (*i.e.* which takes into account the mean-free-path correction in the superconducting state)

$$\lambda = \frac{c}{2\pi\nu_p^*}\sqrt{1+\frac{\xi}{l}}, \tag{2}$$

where l and ξ are the mean free path and the coherence length, respectively. Using the values $\lambda = 4000$–6500 Å and $l/\xi \sim 10$ quoted in ref. [5] and [13], we obtain $h\nu_p^* = 0.31$–0.49 eV, which is in fair agreement with our estimate from the renormalized Drude fit of $\sigma_1(\nu)$.

We can also compare our value of the renormalized plasma frequency $h\nu_p^*$ at 6 K (*i.e.* well within the AFM regime) with the specific-heat coefficient $\gamma(T \to 0) = 150$ mJ/K^2 mole [5], in order to calculate the enhancement of the effective mass m^* and the total charge carrier concentration. From $\omega_p \propto (n/m^*)^{1/2}$ and $\gamma \propto m^* n^{1/3}$, we obtain $m^* = 68 m_e$ and $n_e = 1.09 \cdot 10^{22}$ cm^{-3}. Our estimate of n_e and m^* is also in agreement with the evaluation of these parameters in ref. [5]. The value of n_e corresponds in the case of $m_b = m_e$ to a «normal state» (*i.e.* at $T \gg T_{co}$) plasma frequency of $h\nu_p = 4$ eV. (Indeed, at 300 K and also at 20 K we have fitted our optical conductivity, decoupled from the huge mid-IR absorption at 2000 cm^{-1}, with $h\nu_p = 4$ eV.) We can now apply sum rule or spectral-weight arguments, which, with the assumption that the total charge carrier concentration does not change below T_{co} and above all below T_N (*i.e.* there is *not* any SDW gap at the Fermi surface, which would remove a fraction of the free quasi-particles!), leads to the expression

$$\left(\frac{\nu_p}{\nu_p^*}\right) = \sqrt{\frac{m^*}{m_b}}. \tag{3}$$

We obtain $m^*/m_b \sim 67$ at 16 K and $m^*/m_b \sim 27$ at 15 K, in good agreement with previous evaluations [5].

In conclusion, we have measured the complete electrodynamic response of UPd_2Al_3 at temperatures below T_{co}, and above all in the temperature regime where the AFM transition at 14 K takes place. We find that $\sigma_1(\nu)$ can be described as that of a heavy quasi-particle both somewhat above and below T_N. We have extracted several intrinsic parameters (m^* and τ^*) characterising this coherent many-body state, and found that the enhancement of the effective mass of the heavy quasi-particle is in full agreement with the thermodynamic results.

Our experiments do not give evidence for a SDW energy gap. This is in accordance with the *localised* character of the antiferromagnetic order [4, 5, 13, 14]. We conclude that in UPd_2Al_3 the magnetic ground state develops by interactions other than electron-electron interactions related to the Fermi surface. In this regard, UPd_2Al_3 is fundamentally different from URu_2Si_2, where the opening of a SDW gap is indicative of the *itinerant* nature of the antiferromagnetic order [3].

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REFERENCES

[1] MAPLE M. B., CHEN J. W., DALICHAOUCH Y., KOHARA T., ROSSEL C. and TORIKACHVILI M. S., *Phys. Rev. Lett.*, 56 (1986) 185.
[2] PALSTRA T. T. M., MENOVSKY A. A. and MYDOSH J. A., *Phys. Rev. B*, 33 (1986) 6527.
[3] BONN D. A., GARRETT J. D. and TIMUSK T., *Phys. Rev. Lett.*, 61 (1988) 1305.
[4] GEIBEL C., THIES S., KACZOROWSKI D., MEHNER A., GRAUEL A., SEIDEL B., AHLHEIM U., HELFRICH R., PETERSEN K., BREDL C. D. and STEGLICH F., *Z. Phys. B*, 83 (1991) 305.
[5] GEIBEL C., SCHANK C., THIES S., KITAZAWA H., BREDL C. D., BOEHM A., RAU M., GRAUEL A., CASPARY R., HELFRICH R., AHLHEIM U., WEBER G. and STEGLICH F., *Z. Phys. B*, 84 (1991) 1.
[6] DALICHAOUCH Y., DE ANDRADE M. C. and MAPLE M. B., *Phys. Rev. B*, 46 (1992) 8671.
[7] BARKER jr. A. S., HALPERIN B. I. and RICE T. M., *Phys. Rev. Lett.*, 20 (1968) 384.
[8] BAKKER K., DE VISSER A., TAI L. T., MENOVSKY A. A. and FRANSE J. J. M., *Solid State Commun.*, 86 (1993) 497.
[9] SULEWSKI P. E., SIEVERS A. J., MAPLE M. B., TORIKACHVILI M. S., SMITH J. L. and FISK Z., *Phys. Rev. B*, 38 (1988) 5338.
[10] MARABELLI F. and WACHTER P., *Phys. Rev. B*, 42 (1990) 3307.
[11] FERRERIA I. B., AWASTHI A. M., DEGIORGI L., GRÜNER G., DALICHAOUCH Y. and MAPLE M. B., *Solid State Commun.*, 83 (1992) 27.
[12] AWASTHI A., DEGIORGI L., GRÜNER G., DALICHAOUCH Y. and MAPLE M. B., *Phys. Rev. B*, 48 (1993) to be published.
[13] UEMURA Y. J. and LUKE G. M., *Physica B*, 186-188 (1993) 223.
[14] CASPARY R., HELLMANN P., KELLER M., SPARN G., WASSILEW C., KOEHLER R., GEIBEL C., SCHANK C., STEGLICH F. and PHILLIPS N. E., *Phys. Rev. Lett.*, 71 (1993) 2146.

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1-Loop Analysis of the Photon Self-Energy Due to 3D-Gravity.
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On the Scaling of the Velocity and Temperature Structure Functions in Rayleigh Benard Convection.
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^{13}C-Knight Shift of the Doped Fullerene K_3C_{60}.
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Energetics and Structure of Ordered Sb **Overlayers and** Sb **Clusters on** GaAs(110) **Probed by** *Ab Initio* **Calculations.**
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