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PHYSICA B

Optical investigation of the antiferromagnetic phase transitions in heavy-electron compounds

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Abstract

We have investigated the electrodynamic response of the heavy-electron compounds UPd_2Al_3 ($T_N = 14.5$ K) and UCu_5 ($T_N = 15$ K). The complete excitation spectra were obtained by Kramers–Kronig transformation of the optical reflectivities measured on a fairly broad frequency range (i.e. from 15 up to 10^5 cm^{-1}), combined with the direct measurement of the conductivity at millimeter and microwave frequencies. In UCu_5 we found an absorption in the far infrared below T_N, which is ascribed to excitations across a spin-density-wave-type gap and indicates the itinerant nature of the magnetic phase transition. This feature is absent in UPd_2Al_3, in accordance with the local magnetic moment character of the antiferromagnetic order.

The electrodynamics of several heavy fermion (HF) metals have been explored in detail. However, significantly less is known about the electrodynamics of the magnetically ordered states of these materials. Although magnetic-ordering and heavy-electron behaviour seem, at first sight, to be mutually exclusive, various experimental observations in recent years indicate that this is not necessarily so. Both magnetic ordering out of a heavy-electron state and the formation of a heavy-electron state in a magnetically ordered matrix seem possible. Examples of these two distinctly different situations are realized in the low-temperature properties of U_2Zn_{17}, URu_2Si_2, UPd_2Al_3 and UCu_5, which order antiferromagnetically at 9.7 K, 17 K, 14.5 K and 15 K, respectively. In this paper, we contrast our optical investigations on UCu_5 and on UPd_2Al_3 with the goal of understanding the effects of the magnetic phase transitions in more depth.

For our optical investigations we used well-annealed polycrystalline samples of UCu_5 and single crystals of UPd_2Al_3. We have performed optical reflectivity measurements on a very broad frequency range, from the ultraviolet (UV) down to the far infrared (FIR) and as a function of temperature, as described in previous works [1,2]. Figs. 1(a) and 2(a) summarize the experimental findings in the FIR part of the spectrum, which is relevant for the present discussion. The complete reflectivity spectra were already presented elsewhere [1,2] and particularly our new optical measurements on UPd_2Al_3 single crystals, presented here, confirm the general trend indicated by previous investigations on polycrystalline specimens [1]. A full account of these results will be presented in a forthcoming publication [3]. The optical conductivity $\sigma_1(\omega)$, shown in Figs. 1(b) and 2(b), is obtained through Kramers–Kronig (KK) transformations of the $R(\omega)$ spectra. The spectra were extended to higher frequencies with the usual extrapolations [1,2]. The very delicate low-frequency extrapolation, below the lowest measurable frequency towards zero, was per-

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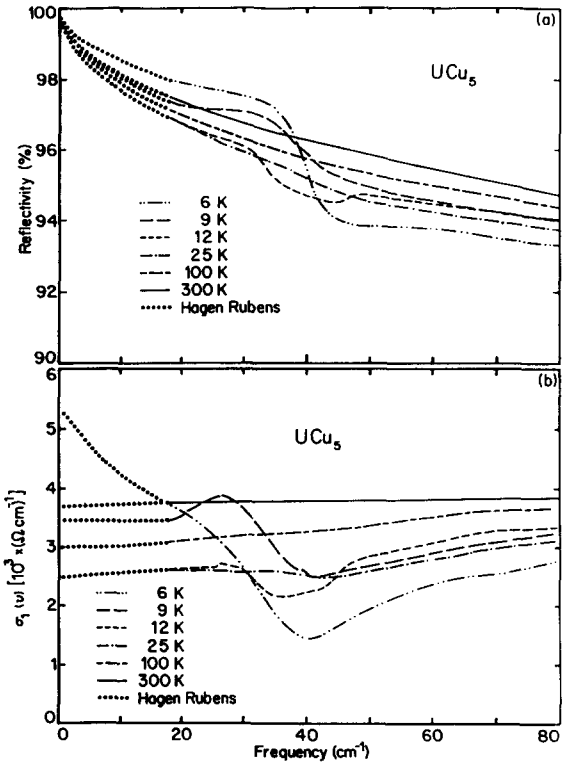

Fig. 1. (a) Reflectivity and (b) optical conductivity in FIR at several temperatures above and below $T_N = 15$ K for UCu_5.

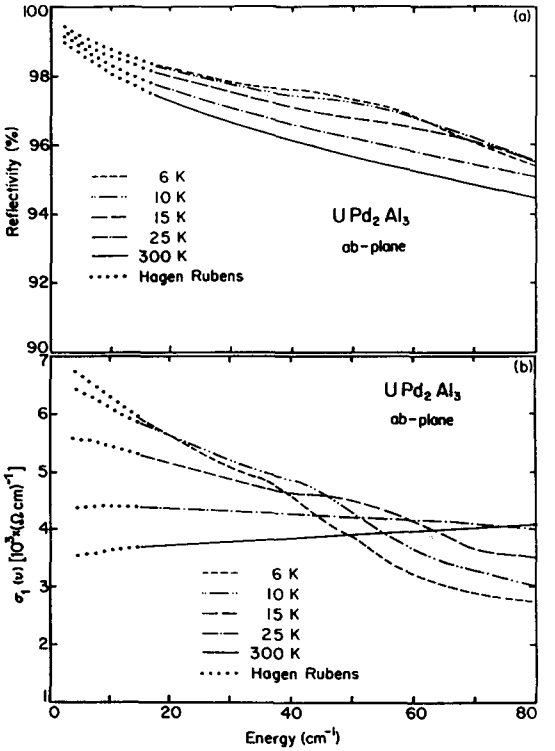

Fig. 2. (a) Reflectivity and (b) optical conductivity in FIR at several temperatures above and below $T_N = 14.5$ K for UPd_2Al_3 single crystals. The light was polarized parallel to the *ab*-plane. The results with light polarized along the hexagonal *c*-axis are similar.

formed with the help of the Hagen–Rubens (HR) law [1,2].

First, we discuss the electrodynamic response of UCu_5, where the most remarkable feature is the fairly strong temperature dependence of $R(\omega)$ in FIR, and particularly below T_N. Indeed, at 25 K the reflectivity has typical metallic behaviour, while at 12 K, where a maximum of $\rho(T)$ is observed [4], we note a deviation from the usual metallic Drude-like behaviour in $R(\omega)$ at about 40 cm^{-1}. As shown in Fig. 1(a), the magnitude of this anomaly grows with decreasing temperature. For a chosen frequency range around 30 cm^{-1} the temperature dependence of $R(\omega)$ produces a very weak bump at 12 K, a well-defined absorption at 9 K and a damped shoulder at 6 K in $\sigma_1(\omega)$ (Fig. 1(b)). We consider it important to note that the general behaviour of $\sigma_1(\omega)$ in the measured frequency range is completely unaffected by the HR extrapolation. Moreover, it is worth noting that the electrodynamic response of UCu_5 bears several similarities with the optical properties of URu_2Si_2 [5].

We claim that the temperature dependence of $\sigma_1(\omega)$ in FIR of UCu_5 is related to the onset of the antiferromagnetic order below T_N. The absorption most clearly seen at 9 K is interpreted as being due to a spin density wave (SDW) gap [6], consequently implying the partly itinerant nature of the antiferromagnetic phase transition at T_N. The optical conductivity below T_N can consistently be fitted by a combination of two contributions, namely a low-frequency 'renormalized' Drude contribution, and a phenomenological harmonic oscillator for the absorption at about 30 cm^{-1}. Based on these assumptions we may then evaluate the resonance frequency which we ascribe to excitations across the SDW gap. Its saturation value is 28 cm^{-1} and corresponds to a reduced gap of $2\Delta/k_B T_N = 2.7$. It is remarkable that this ratio is in agreement with a previous evaluation arrived at by an analysis of the anomaly in $\rho(T)$ around T_N, assuming a two-component description of the total conductivity [4].

On the other hand, UPd_2Al_3 does not display a

similar absorption, the onset of which would be coincident with T_N. In fact, we do not find a feature at (or near to) $2\Delta = 3.52k_B T_N$, which could be ascribed to a so-called SDW gap. This peculiar behaviour of $\sigma_1(\omega)$ 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 DC 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 in the relaxation time τ [1]. Similar results were also obtained for U_2Zn_{17} [2]. The absence a SDW energy gap is in accordance with the localised character of the antiferromagnetic order [7].

The temperature dependence in $\sigma_1(\omega)$ is due to a temperature dependent narrow Drude-like resonance in the FIR range, ascribed to the optical conductivity of the heavy quasiparticle. This behaviour is reminiscent of what has been found in other HF systems, like, for example, in $CeAl_3$ [8]. The narrow resonance centred at $\omega = 0$ can be fitted, again, with the so-called renormalized Drude expression [8], which leads to the values $m^*/m_b \sim 67$ at 6 K and $m^*/m_b \sim 27$ at 15 K for the effective mass, in good agreement with evaluations from the specific heat data [1,9].

In conclusion, we have shown that the electrodynamic response in UCu_5 and UPd_2Al_3 is indicative of the different nature of the antiferromagnetic phase transitions, observed in these compounds, which also seems to be reflected in the corresponding different behaviours of $\rho(T)$. Nevertheless, it remains to be seen what kind of relation might be established between typical itinerant features in the electrodynamic response of these antiferromagnets, particularly of UCu_5 and URu_2Si_2, and their rather simple magnetic order, apparently commensurate with the lattice.

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