EUROPHYSICS LETTERS 20 April 1994

Europhys. Lett., 26 (3), pp. 221-226 (1994)

Optical Probing of the Antiferromagnetic Phase Transitions in the Heavy-Electron Compounds U_2Zn_{17} and UCu_5.

L. DEGIORGI(*), H. R. OTT(*), M. DRESSEL(**)
G. GRÜNER(**) and Z. FISK(***)

(*) *Laboratorium für Festkoerperphysik, ETH-Zürich*
CH-8093 Zürich, Switzerland
(**) *Department of Physics, University of California at Los Angeles*
Los Angeles, CA 90024, USA
(***) *Materials Science and Technology Division, Los Alamos National Laboratory*
Los Alamos, NM 87545, USA

(received 21 January 1994; accepted 1 March 1994)

PACS. 78.20 – Optical properties of bulk materials.
PACS. 71.28 – Narrow-band systems, heavy-fermion metals: intermediate-valence solids.

Abstract. – We have investigated the complete electrodynamic response of the heavy-electron compounds U_2Zn_{17} and UCu_5. Particular emphasis has been devoted to the optical evidence of the antiferromagnetic phase transitions at 9.7 K and 15 K for U_2Zn_{17} and UCu_5, respectively. In UCu_5, we found an absorption in the far infrared, which is ascribed to excitations across a spin-density-wave–type gap. This feature is absent in U_2Zn_{17}. We argue that UCu_5 belongs to a characteristically different class of antiferromagnets than U_2Zn_{17} which represents the class of heavy-electron compounds with localized magnetic moments.

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 necessary 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} [1] and UCu_5 [5], which order antiferromagnetically at 9.7 K and 15 K, respectively. Neutron diffraction experiments reveal rather conventional types of arrangements of the ordered moments, in both cases of the order of $1\mu_B$ per U ion [3-5]. These phase transitions are indicated by distinct anomalies of the specific heat and abrupt changes in the temperature dependence of the magnetic susceptibility. The ordering temperatures may also be identified by non-monotonous variations of the temperature dependence of the electrical resistivity. For U_2Zn_{17}, the positive slope $\delta\rho/\delta T$ suddenly increases with decreasing temperature, manifesting lesser scattering of conduction electrons at moment fluctuations. For UCu_5 the behaviour is distinctly different [2, 6]. The slope $\delta\rho/\delta T$ is negative in the paramagnetic state. Within a small temperature interval it displays a tendency towards divergence but finally changes discontinuously to a large negative value, *i.e.* $\rho(T)$ increases considerably with

decreasing temperature just below T_N. Decaying magnetic scattering and the onset of coherence as the heavy-electron state develops well below T_N result in a maximum of $\rho(T)$ approximately 3 K below the Néel temperature. Nevertheless, the $\rho(T)$ variation close to but below T_N implies that the energy spectrum of the itinerant charge carriers is considerably affected by the phase transition, in contrast to what is observed in U_2Zn_{17} [1].

In this paper we report on a, to our knowledge, first attempt to study these features in more depth by probing the effects of the phase transitions with optical methods. From our observations we conclude that U_2Zn_{17} undergoes an antiferromagnetic phase transition involving mainly local magnetic moments. In contrast, UCu_5 exhibits an excitation spectrum characterized by the sudden appearance of absorption in the far-infrared (FIR) spectral range at a temperature coincident with T_N, which we tend to ascribe to a SDW gap. However, our experimental results on UCu_5 provide a rather puzzling contrast between the SDW-like picture emerging from our optical work and the commensurability of the magnetic order [3, 4]. In principle, optical experiments are a very suitable tool because an *incommensurate* antiferromagnetic order should lead to a spin density wave (SDW) formation and an opening of a gap at the Fermi level ε_F, leading to a distinct absorption in the excitation spectrum at $2\Delta = 3.52 k_B T_N$. Such a SDW arises as a result of a Fermi-surface instability, due to the degeneracy between occupied levels on one side and empty levels on the other side of the Fermi-distribution edge. In the case of a *commensurate* antiferromagnetic order we would not expect a gap at ε_F for partially filled bands. In fact, the degeneracy would be destroyed by the periodic crystal potential with large band structure gaps, except at the unlikely situation where the latter potential is very small [7].

Well-annealed polycrystalline samples of U_2Zn_{17} and UCu_5 have been used for our optical investigations. Previous experiments on U_2Zn_{17} have shown that all features of low-temperature properties are independent of whether they are measured on poly- or single-crystalline material. For UCu_5 we have investigated the same specimen, previously used for studies of low-temperature transport properties [6]. We have performed optical-reflectivity measurements on a very broad frequency range, from the ultraviolet (UV) down to the far infrared (FIR) and, in both cases, as a function of temperature. To this end, we used four spectrometers with overlapping frequency ranges, as described in previous work [8]. Both samples were polished in order to obtain flat and shiny surfaces.

In fig. 1 we present the reflectivity spectra on the whole measured frequency range for both compounds at 300 K (note the logarithmic frequency scale). The insets display the $R(\omega)$ spectra at several temperatures above and below T_N in the FIR range (note the linear frequency scale). At frequencies above the mid-IR there is no temperature dependence discernible in the reflectivity spectra of both compounds. The optical conductivity $\sigma_1(\omega)$ is obtained through Kramers-Kronig (KK) transformations of the $R(\omega)$ spectra. The spectra were extended to higher frequencies with the usual extrapolations $R(\omega) \sim 1/\omega^2$ for $\omega < 3 \cdot 10^5\,\mathrm{cm}^{-1}$ and $R(\omega) \sim 1/\omega^4$ for higher frequencies. The very delicate low-frequency extrapolation, below the lowest measurable frequency (*i.e.* $17\,\mathrm{cm}^{-1}$) towards zero, was performed with the help of the Hagen-Rubens (HR) relation $R(\omega) = 1 - 2\sqrt{\omega/\sigma_{\mathrm{d.c.}}}$, using the measured $\sigma_{\mathrm{d.c.}}$ data [1, 2, 6]. We discuss this in more detail below.

Figure 2 displays the complete excitation spectra in terms of the optical conductivity on a logarithmic frequency scale. The FIR part is shown in fig. 3 for several temperatures. The excitation spectra in the mid-IR frequency range are characterized by a dominant absorption, centered at 3000 and $4000\,\mathrm{cm}^{-1}$ for UCu_5 and U_2Zn_{17}, respectively. At higher frequencies, *i.e.* in the ultraviolet, some weak structures may be identified. A complete band structure calculation would be of help for a precise identification of these absorptions; we tend to ascribe them to electronic interband transitions.

First, we discuss the electrodynamic response of UCu_5, with particular emphasis on the

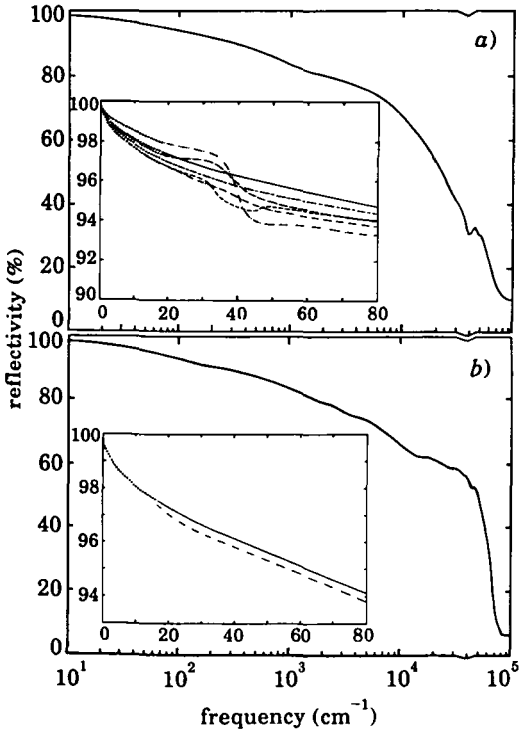

Fig. 1.

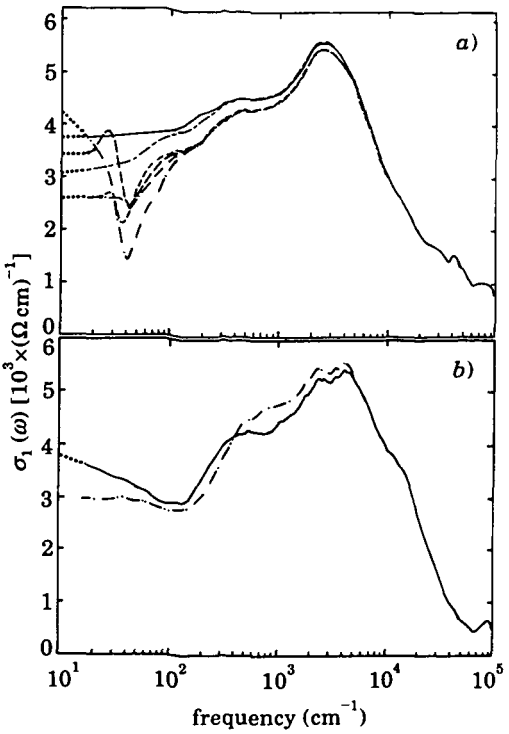

Fig. 2.

Fig. 1. – Room temperature reflectivity spectra of UCu_5 (*a*)) and U_2Zn_{17} (*b*)) (note the logarithmic energy scale). The insets display a blow-up of the FIR energy range (linear energy scale) at several temperatures: ··–·· 6 K, —— 9 K, --- 12 K, –·– 25 K, –·– 100 K, —— 300 K, ···· Hagens Rubens.

Fig. 2. – Optical conductivity at several temperatures (meaning of the curves is as in fig. 1) for UCu_5 (*a*)) and U_2Zn_{17} (*b*)) in the whole frequency range (note the logarithmic energy scale).

FIR spectral range. The most remarkable feature is the fairly strong temperature dependence of $R(\omega)$ in FIR. The inset of fig. 1*a*) displays this frequency range, of particular relevance for the present discussion. It may be seen that the variation of the temperature dependence of $R(\omega)$ is considerable below T_N. Indeed, at 25 K the reflectivity has the typical metallic behaviour, while at 12 K, where a maximum of $\rho(T)$ is observed [2, 6], we note a deviation from the usual metallic Drude-like behaviour in $R(\omega)$ at about 40 cm^{-1}. As shown in the inset of fig. 1*a*), the magnitude of this anomaly grows with decreasing temperature. It is interesting to observe that, nevertheless, the lowest part of $R(\omega)$ (fig. 1*a*)) at each temperature matches very well a HR extrapolation to 100% reflectivity at $\omega = 0$ and taking into account the previosly measured $\sigma_{d.c.}(T)$ values [6]. Considering fig. 3*a*), one may also check the fairly good correspondence between the FIR conductivity, *i.e.* $\sigma_1(\omega \to 0)$ limit, and the measured $\sigma_{d.c.}$ values of this sample [6], which calls for the good consistency of the KK transformations.

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. 3*a*)). The low-frequency optical conductivity, which is merely the consequence of the HR extrapolation, can be described with a renormalized Drude

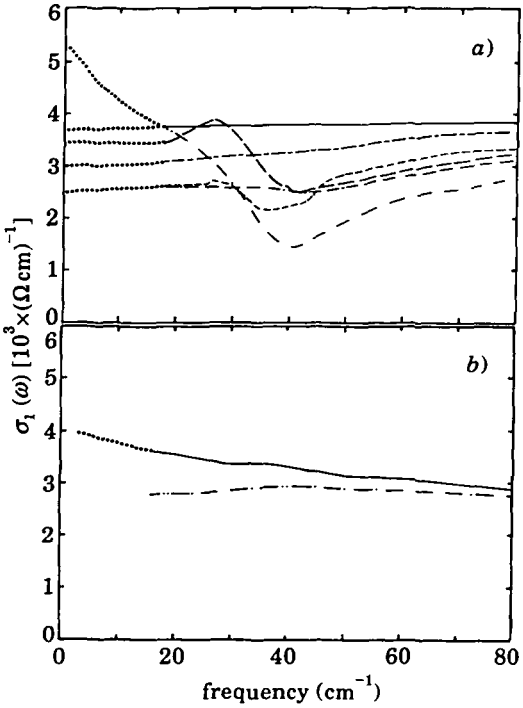

Fig. 3. – Optical conductivity at several temperatures (meaning of the curves is as in fig. 1) in the FIR energy range (linear energy scale) for UCu_5 (*a*)) and U_2Zn_{17} (*b*)).

behaviour, defined by enhanced effective mass m^* and relaxation time τ^*. We shall return to this aspect later. 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. It is well known that the evaluation of this part of the spectrum is fairly delicate and that $\sigma_1(\omega)$ may be strongly altered by ways of different extrapolations. We checked this aspect by performing the KK transformations without the extension of the spectra in the form of the HR extrapolation. The bump or absorption at about 30 cm^{-1} in $\sigma_1(\omega)$ turns out to be even more enhanced in that case. Thus, we consider the performed extrapolation as a safe approach for the analysis of the optical spectra, leading therefore to a rather cautious interpretation.

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 SDW gap [7], 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}. Details of the phenomenological fit will be presented elsewhere. 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 $\rho(T)$ around T_N, assuming a two-component description of the total conductivity [6]. This latter analysis also suggests that only 20% of the total charge carrier concentration above T_N is involved in the SDW phase transition. Hence, the remaining free charge carriers (*i.e.* ~ 80%) contribute to the optical conductivity with

the low-frequency Drude behaviour. This contribution overlaps the SDW gas absorption. Indeed at 6 K, the SDW gap appears at best as a shoulder. At 6 K, $\sigma_{d.c.}$ is two times larger than at T_N and the renormalized Drude contribution partially smears the SDW gap absorption. There is a compelling similarity with the optical properties of URu_2Si_2 [9] and UNi_2Si_2 [10], where, in analogy to the situation in Cr [11], a SDW gap was also identified.

We now compare the electrodynamic response of UCu_5 with that of U_2Zn_{17}. As we have already mentioned above, we find no appreciable variation of the temperature dependence of $R(\omega)$ in FIR when crossing the antiferromagnetic phase transition temperature of U_2Zn_{17} with decreasing temperature. Down to our lowest frequency (*i.e.* 17 cm^{-1}) there is no indication of an absorption appearing below T_N (fig. 3*b*)). Unless an eventual SDW gap absorption develops at very and thus anomalously low frequencies, we tend to ascribe U_2Zn_{17} to the class of materials with an antiferromagnetic phase transition involving local magnetic moments. Nevertheless, it cannot *a priori* be excluded that this localized antiferromagnetic order still opens gaps in the electronic band structure, due to the crystal potential and depending on the degree of interaction between possible new boundaries of the Brillouin zone (BZ) and the Fermi surface (*i.e.* depending on whether the zone boundaries only touch or cut the Fermi surface) [12]. However, this possibility is very unlikely since the magnetic BZ of U_2Zn_{17} has been claimed to be identical with the chemical zone [5].

The electrodynamic response of U_2Zn_{17} is reminiscent of our recent experimental findings on UPd_2Al_3 [13]. In U_2Zn_{17}, as indicated by fig. 2*b*) and particularly by fig. 3*b*), the $\omega \to 0$ limit of $R(\omega)$ and, above all, of $\sigma_1(\omega)$ at low temperatures would be consistent with a HR extrapolation with a very low $\sigma_{d.c.}$ value. This is, however, in contrast with the $\rho(T)$ results. Therefore, we performed a measurement of the surface resistance R_s at 35 and 100 GHz with the cavity perturbation technique [8]. One can then calculate σ_1 (fig. 4) from R_s with the assumption that $X_s = R_s$, X_s being the surface reactance. Even though the latter assumption might be too crude [8], it is, nevertheless, gratifying that at low temperatures we obtain a fairly strong frequency dependence of $\sigma_1(\omega)$, with the development of a narrow resonance centered at zero frequency. As in UPd_2Al_3 [13], a fit with the usual renormalized Drude model can fully describe this part of the excitation spectrum. The small amount of spectral weight associated with the narrow renormalized Drude resonance is indicative of an

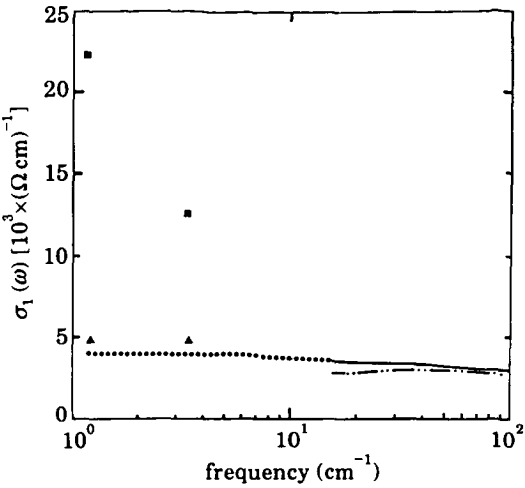

Fig. 4. – Optical conductivity of U_2Zn_{17} in the FIR frequency range at 300 (——) and 6 K (·—·) together with the measurements at 35 and 100 GHz: ■ at 6 K, ▲ at 300 K.

enhancement of the effective mass of the quasi-particles, in agreement with the thermodynamic results [1].

In conclusion, we have shown that the electrodynamic response in UCu_5 and U_2Zn_{17} is indicative of the different nature of the antiferromagnetic phase transitions observed in these compounds. There is, however, an interesting analogy between the excitation spectrum of UCu_5 and URu_2Si_2, and between that of U_2Zn_{17} and UPd_2Al_3, which 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_3, and their rather simple magnetic order, apparently commensurate with the lattice.

* * *

The authors wish to thank P. WACHTER for important financial support, T. M. RICE, R. HLUBINA and R. MONNIER for helpful discussions, and J. MÜLLER for technical assistance. The work is also part of a joint collaboration under the auspices of a grant by the Swiss-US National Foundations for the Scientific Research. One of us (MD) acknowledges the financial support from the Alexander von Humboldt Foundation.

REFERENCES

[1] OTT H. R., RUDIGIER H., DELSING P. and FISK Z., *Phys. Rev. Lett.*, 52 (1984) 1551.
[2] OTT H. R., RUDIGIER H., FELDER E., FISK Z. and BATLOGG B., *Phys. Rev. Lett.*, 55 (1985) 1595.
[3] MURASIK A., LIGENZA S. and ZYGMUNT A., *Phys. Status Solidi A*, 23 (1974) K163.
[4] SCHENCK A., BIRRER P., GYGAX F. N., HILTI B., LIPPELT E., WEBER M., BOENI P., FISCHER P., OTT H. R. and FISK Z., *Phys. Rev. Lett.*, 65 (1990) 2454.
[5] COX D. E., SHIRANE G., SHAPIRO S. M., AEPPLI G., FISK Z., SMITH J. L., KJEMS J. and OTT H. R., *Phys. Rev. B*, 33 (1986) 3614.
[6] BERNASCONI A., MOMBELLI M., FISK Z. and OTT H. R., to be published in *Z. Phys. B*.
[7] OVERHAUSER A. W., *Phys. Rev.*, 128 (1962) 1437.
[8] AWASTHI A. M., DEGIORGI L., GRÜNER G., DALICHAOUCH Y. and MAPLE M. B., *Phys. Rev. B*, 48 (1993) 10692.
[9] BONN D. A., GARRETT J. D. and TIMUSK T., *Phys. Rev. Lett.*, 61 (1988) 1305.
[10] CAO N., GARRETT J. D. and TIMUSK T., *Physica B*, 191 (1993) 263.
[11] BARKER jr. A. S., HALPERIN B. I. and RICE T. M., *Phys. Rev. Lett.*, 20 (1968) 384.
[12] ELLIOTT R. J. and WEDGWOOD F. A., *Proc. Phys. Soc.*, 81 (1963) 846.
[13] DEGIORGI L., DRESSEL M., GRÜNER G., WACHTER P., SATO N. and KOMATSUBARA T., *Europhys. Lett.*, 25 (1994) 311.