Nature of Heavy Quasiparticles in Magnetically Ordered Heavy Fermions UPd_2Al_3 and UPt_3

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The optical conductivity of the heavy fermions UPd_2Al_3 and UPt_3 has been measured in the energy range from 0.04 to 5 meV. In both compounds a well pronounced pseudogap of less than 1 meV develops in the optical response at low temperatures; we relate this to the antiferromagnetic ordering. From the energy dependence of the effective mass and scattering rate we conclude that the enhancement of the mass mainly occurs below the energy which is related to magnetic correlations between the local magnetic moments and the itinerant electrons. This implies that the magnetic order in these compounds is the prerequisite to the formation of the heavy quasiparticles and eventually to superconductivity.

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In common metals itinerant electrons of the conduction band are responsible for the electrical transport. The conductivity is constant for frequencies up to the scattering rate Γ. This behavior is well described by the Drude model: $\hat{\sigma}(\omega) = \sigma_1 + i\sigma_2 = (Ne^2/m)/(\Gamma - i\omega)$ with the charge carrier mass m and scattering rate Γ assumed to be frequency independent. Here N is the carrier concentration and e is the electronic charge [1]. While at room temperature heavy fermions (HF) also follow this scenario; at liquid-helium temperatures large deviations are observed, which are caused by many-body effects.

Heavy fermions show an interplay of electronic correlations due to an interaction of itinerant electrons and local magnetic moments. At low temperatures these intermetallic compounds exhibit a significant increase of the magnetic susceptibility and the electronic contribution to the specific heat as compared to most metals. This is commonly explained by an enhanced effective mass m^* of some hundred times the free electron mass due to the highly correlated electronic behavior and is described in the context of interacting Fermi liquids [2]. The strong interaction of the quasi-free conduction-band electrons with nearly localized f electrons leads to a many-body resonance, i.e., an enhanced density of states (DOS) at the Fermi energy [3]. The growth of this narrow resonance sets in below the coherence temperature T^* (typically 10 to 100 K), which is accompanied by a drop in the dc resistivity and a broad maximum in the magnetic susceptibility.

Also the optical properties show clear signatures of the coherent many-body ground state: As a consequence of the large effective mass m^* the spectral weight $\int_0^{\omega_c} \sigma_1(\omega)\, d\omega = \omega_p^{*2}/8$ is reduced. ω_c is a cutoff frequency which makes sure that only the free electrons are considered; $\omega_p^* = (\frac{4\pi Ne^2}{m^*})^{1/2}$ is the renormalized plasma frequency. The relevant electronic excitations shift to very low energies, best characterized by an effective scattering rate Γ^* [4]. With decreasing temperature the electronic correlations become more pronounced, leading to a gradual increase of m^* and the corresponding decrease of Γ^*. The interaction of the conduction electron spins with the atomic moments in the heavy-fermion crystals explains the reduction of the magnetic susceptibility and also leads to a hybridization gap Δ in the DOS. The gap scales with the coherence temperature $(\Delta/T^*)^2 = m/m^*$ and is commonly observed in the far infrared spectral range [5,6]. Using a frequency dependent scattering rate and an effective mass for the description of the optical properties of correlated electron systems one obtains the energy dependence of the renormalization effects which goes beyond the information derived from static thermodynamic measurements. In the low-energy limit the frequency dependence of both parameters $m^*(\omega)$ and $\Gamma^*(\omega)$ should resemble their temperature dependence [1].

An additional aspect becomes important for those HF compounds which undergo a magnetic phase transition. Antiferromagnetism which is due to the ordering of localized magnetic moments does not lead to a gap at the Fermi energy since the order is commensurate with the lattice. In the case of a spin density wave, a spatial modulation of the conduction-electron spins leads to antiferromagnetism, which in general is incommensurate with the underlying lattice; a gap in the electronic DOS at the Fermi energy is observed. Optical experiments exhibit a clear gap feature in the latter case, while for the first case we expect no influence on the optical properties [7].

Because of its two-component electronic character, UPd_2Al_3 exhibits both a pronounced local magnetic moment and HF itinerant behavior [8–10]. The $5f$ shell has an average occupation of slightly less than 3, with two electrons localized in the U^{4+} state and the remaining $5f$ electron considered to be itinerant due to their large hybridization with the conduction electrons [11]. This coexistence is seen in the large magnetic moment ($0.85\mu_B$) well below the antiferromagnetic ordering temperature

$T_N \approx 14$ K which is almost atomiclike, while at the same time the itinerant electrons display a large coefficient of the electronic specific heat $\gamma = 140\ \mathrm{mJ\,mol^{-1}\,K^{-2}}$ which indicates $m^*/m \approx 50$. The magnetic ordering is of local origin and the formation of a spin density wave can be ruled out [7,12]. The heavy quasiparticles condense in the superconducting state below $T_c \approx 2$ K. In order to reconcile magnetic ordering and superconductivity it was first suggested that different parts of the Fermi surface lead to antiferromagnetism and to the HF ground state. Recent observations by tunneling spectroscopy and inelastic neutron scattering indicate, however, that magnetic excitons produce the effective coupling between the itinerant electrons and are responsible for superconductivity in UPd_2Al_3 [13,14].

If there is an interaction between magnetic and electronic excitations in the superconducting state, the question arises as to whether the magnetic ordering also affects the heavy quasiparticles in the normal state. To which degree is the low-temperature ground state (still $T > T_c$) with the large m^* affected by the magnetic ordering? Are the correlations of the electrons in the f shell with those in the conduction band solely responsible for this mass enhancement, as was argued within the common HF picture?

In order to address these questions, we have investigated the optical properties of UPd_2Al_3 in the frequency range 10 GHz–1.2 THz (i.e., 0.3–40 $\mathrm{cm^{-1}}$ or 0.04–5 meV) at temperatures $2 < T < 300$ K. Using a Mach-Zehnder interferometer equipped with tunable, coherent radiation sources we measured the transmission and the phase shift of the radiation upon passing through a 150 nm thick UPd_2Al_3 film deposited on a $LaAlO_3$ substrate by molecular beam epitaxy [15]. In addition we performed microwave experiments utilizing enclosed cavities [16]. Our data were supplemented with reflectivity measurements on bulk samples [7] in order to obtain the absorptivity over a broad frequency range as plotted in Fig. 1a. The optical conductivity (Fig. 1b) is obtained by fitting both the absorptivity and the conductivity data, taking the different experimental uncertainties into account [16].

The main findings can be summarized as follows: At high temperatures we observe a broad conductivity spectrum as expected for a Drude metal. As T decreases below $T^* \approx 50$ K, a renormalized Drude peak develops due to the gradual enhancement of m^*. At the same time, a gap feature develops around 100 $\mathrm{cm^{-1}}$ as expected from the hybridization of the localized $5f$ electrons and the conduction electrons [4]. Lowering the temperature further, magnetic ordering sets in at T_N and leads to distinct signatures in the optical spectrum. In the magnetically ordered state we observe a pronounced pseudogap below 2 $\mathrm{cm^{-1}}$ which we assign to the interaction of the localized magnetic moments and the spins of the itinerant electrons. At even lower frequencies, an extremely narrow zero-frequency mode remains which is responsible for the dc conductivity and for superconductivity below T_c.

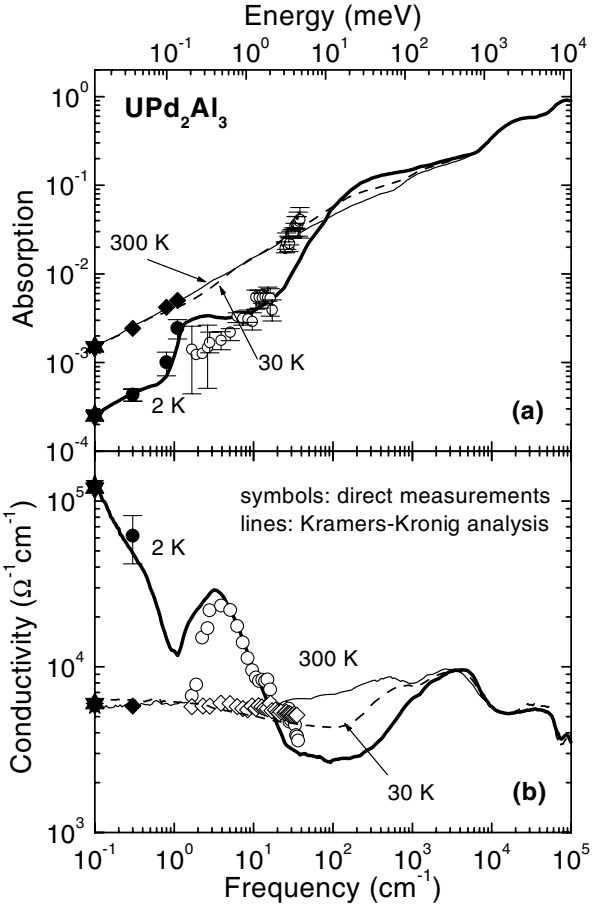

FIG. 1. (a) Frequency dependent absorption of UPd_2Al_3 at different temperatures. The symbols on the left axis represent the dc values in a Hagen-Rubens behavior; the full symbols in the microwave range are obtained by the cavity perturbation technique; the open symbols represent absorption evaluated from the transmission and phase measurement by the Mach-Zehnder interferometer (only 2 K data are plotted for clarity reasons). The lines are obtained by combining the various optical investigations (transmission through films and reflection of bulk samples). (b) Corresponding optical conductivity of UPd_2Al_3 as evaluated by a Kramers-Kronig analysis of the above absorption data. The points on the left axis indicate the dc conductivity; the full symbols correspond to the direct determination of σ_1 using the transmission and the phase shift obtained by the Mach-Zehnder interferometer.

To describe the low-energy data satisfactorily, we introduce a complex frequency dependent scattering rate $\hat{\Gamma}(\omega) = \Gamma_1(\omega) + i\Gamma_2(\omega)$ into the standard Drude form of $\hat{\sigma}(\omega)$. With the dimensionless quantity $\lambda(\omega) = -\Gamma_2(\omega)/\omega$, the complex conductivity can be written as [1]

$$\hat{\sigma}(\omega) = \frac{(\omega_p')^2}{4\pi} \frac{1}{\Gamma_1(\omega) - i\omega[m^*(\omega)/m]}, \qquad (1)$$

where $m^*/m = 1 + \lambda(\omega)$ is the frequency dependent enhanced mass. $(\omega_p')^2$ corresponds to the fraction of electrons which participate in the many-body state below T^*. By rearranging Eq. (1) we get

$$\Gamma_1(\omega) = \frac{(\omega_p')^2}{4\pi} \frac{\sigma_1(\omega)}{|\hat{\sigma}(\omega)|^2}, \quad (2)$$

$$\frac{m^*(\omega)}{m} = \frac{(\omega_p')^2}{4\pi} \frac{\sigma_2(\omega)/\omega}{|\hat{\sigma}(\omega)|^2}. \quad (3)$$

Such an analysis allows us to identify for interactions which would lead to an energy-dependent renormalization as $\Gamma_1(\omega)$ and $m^*(\omega)$ are related to the real and imaginary parts of the energy-dependent self-energy of the electrons [17]. In Fig. 2 the frequency dependences of the scattering rate and effective mass are displayed for different temperatures. As expected for a normal metal, at $T > T^*$ both spectra are nearly frequency independent. As T is lowered we observe a peak in $\Gamma_1(\omega)$ at the hybridization gap, as can be nicely seen in the $T = 30$ K data of Fig. 2b. The decrease of $\Gamma_1(\omega)$ to lower frequencies corresponds to an increase of $m^*(\omega)$. At these intermediate temperatures, $T_N < T < T^*$, the effective mass m^* gradually increases when going to smaller frequencies or correspondingly to lower temperatures; eventually it reaches a value of $35m$. Decreasing the temperature below T_N results in a second peak of $\Gamma_1(\omega)$ and a strong increase of the effective mass below 2 cm^{-1}. As already seen at $T = 15$ K, this effect becomes stronger as the temperature is lowered to 2 K. The effective mass $m^*(\omega)$ levels off below this frequency at $m^*/m \approx 50$ and nicely matches the value obtained by thermodynamic methods (Fig. 2c) [8]. We see a moderate enhancement of $m^*(\omega, T \to 0)$ for frequencies below the hybridization gap and a larger one below the gap which develops due to magnetic correlations. Accordingly $m^*(\omega \to 0, T)$ increases only slightly below T^* and shows a large enhancement below T_N. The scattering rate drops more than 1 order of magnitude upon passing T_N; this can be explained by the freezing out of spin-flip scattering in the ordered state. The energy-dependent scattering rate is related to the electronic DOS. In the case of UPd_2Al_3 we can distinguish two gap structures with an enhanced DOS at the edges, features known for superconductors [16].

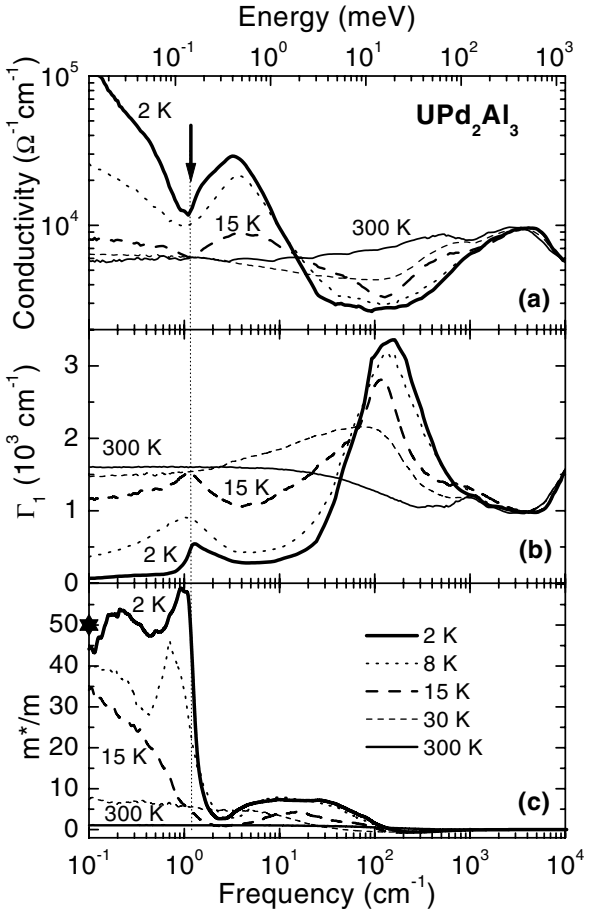

FIG. 2. Frequency dependence of (a) the optical conductivity $\sigma_1(\omega)$, (b) the scattering rate $\Gamma_1(\omega)$, and (c) the effective mass $m^*(\omega)$ of UPd_2Al_3 for different temperatures. The point on the left axis corresponds to m^* derived by thermodynamic measurements [8]. The arrow marks the correlation gap.

The estimation of the superconducting condensate plasma frequency $\omega_p^s = c/\lambda_L \approx 4000$ cm^{-1} (c is the velocity of light) from the London penetration depth $\lambda_L = 450$ nm [9] agrees with our results of $\omega_p^* = 4300$ cm^{-1} if we integrate $\sigma_1(\omega)$ up to $\omega_c = 100$ cm^{-1} [18]; a renormalized Drude behavior without a gap feature would yield a value more than twice as large. This implies that all carriers seen in our low-energy spectra are in the HF ground state and eventually undergo the superconducting transition below T_c. We can definitely rule out an assignment of the gap simply to the localized carriers of the antiferromagnetically ordered state, with the delocalized carriers contributing only to the narrow feature at $\omega = 0$, because its small spectral weight at low temperatures accounts for only 18% of the carriers which become superconducting. This also means that the excitations above the gap stem from the delocalized states and that the gap observed in our spectra is related to exchange correlations of the second subsystem. Recently it was found that the magnetic excitations seen by neutron scattering produce effective interactions between itinerant electrons and lead to superconductivity [14]. Our results imply that the magnetic excitations responsible for superconductivity also cause the heavy quasiparticles.

The picture we developed to explain the optical properties of UPd_2Al_3 may also apply to other HF compounds, in particular, to UPt_3 which exhibits a similar behavior in many regards [2,19]. For UPt_3 the effective mass of the quasiparticles is larger ($m^*/m \approx 200$) and the relevant energy scales are lower. The coherence temperature is $T^* \approx 30$ K, fluctuating short range magnetic order occurs at $T_N = 5$ K, and superconductivity sets in at $T_c = 0.5$ K. Although the magnetic moment ($0.02\mu_B$) is much smaller, recent band structure calculations [20] infer the existence of localized as well as delocalized $5f$ electrons in UPt_3 very similar to UPd_2Al_3. It was suggested that the enhancement of the quasiparticle mass results from the local exchange interaction of two localized $5f$ electrons with the remaining delocalized ones [20].

We have reanalyzed the optical properties of UPt_3 previously obtained [21,22] in the same way as described above

and can identify similar features (Fig. 3). When the coherent ground state builds up ($T < T^*$), the optical conductivity increases for frequencies below 30 cm^{-1}. As T drops below $T_N = 5$ K, an energy gap progressively develops at about 3 cm^{-1} which is assigned to magnetic correlations [22]. The frequency dependence of the effective mass displayed in Fig. 3c clearly shows at the lowest temperature accessible that only a marginal increase of m^* is observed around 30 cm^{-1}, while below the energies related to the magnetic correlations the mass is drastically enhanced. Thus we argue that, as for UPd_2Al_3, the coupling of the localized and delocalized $5f$ electrons causes the heavy quasiparticles in UPt_3; these magnetic excitations are very likely to be responsible for superconductivity.

For both compounds, it appears that the formation of the heavy quasiparticles relies on the establishment of antiferromagnetic order rather than on competition of the coherent singlet formation and the magnetic order. Thermodynamic measurements should test our idea that $m^*(T)$ increases only slightly below T^* but shows a large enhancement below T_N. Since we made similar observations for two materials which may have a different ground state, as evidenced by the vastly different μ_B, it is of interest to test how far our findings have implications on nonmagnetic HF, U based but also Ce based. It also remains to be seen in which way our observation is related to the pseudogap which was established in high-temperature superconductors well above T_c by optical and other methods [23].

In conclusion, the electrodynamic response of two uranium-based heavy fermion superconductors with magnetic ordering exhibits a behavior at low temperatures and low frequencies which cannot be explained within the simple picture of a renormalized Fermi liquid. The low-energy response shows the well-known renormalized behavior only for temperatures above the magnetic ordering; below T_N additional features appear. Besides an extremely narrow zero-frequency response, we observe a pseudogap of less than 1 meV. The experiments indicate that this pseudogap is connected to magnetic correlations between localized and delocalized charge carriers. We argue that this gap at extremely low energies is due to the influence of the localized magnetic moments on the conduction electrons and this interaction is mainly responsible for the enhanced mass m^*.

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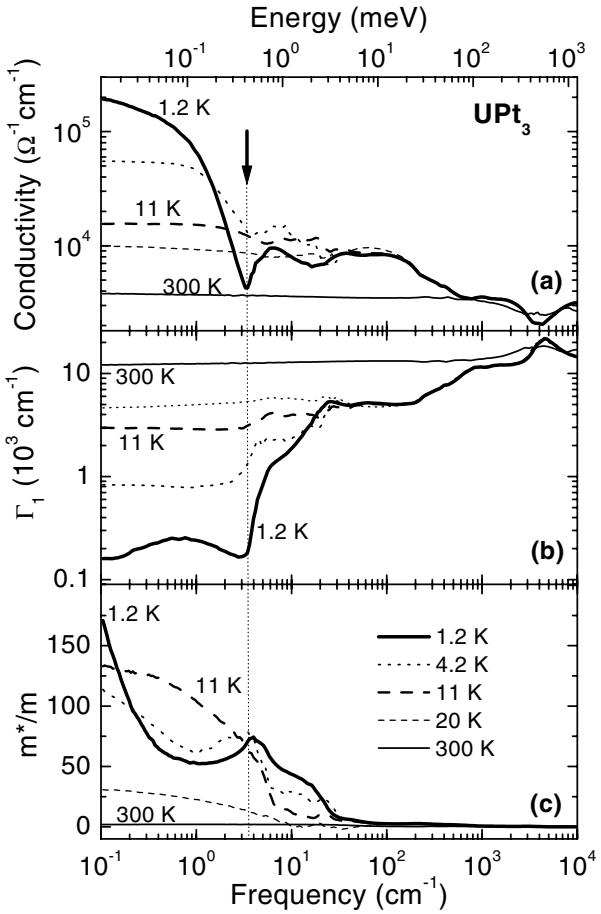

FIG. 3. Frequency dependence of (a) the optical conductivity, (b) the scattering rate, and (c) the effective mass of UPt_3 for different temperatures (optical data from [21,22]).

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[1] M. Dressel and G. Grüner, *Electrodynamics of Solids* (Cambridge University Press, Cambridge, 2002).
[2] Z. Fisk *et al.*, Science **239**, 33 (1988).
[3] N. Grewe and F. Steglich, in *Handbook on the Physics and Chemistry of Rare Earths,* edited by K. A. Gscheidner and L. Eyring (Elsevier, Amsterdam, 1991), Vol. 14, p. 343.
[4] L. Degiorgi, Rev. Mod. Phys. **71**, 687 (1999).
[5] A. C. Hewson, *The Kondo Problem to Heavy Fermions* (Cambridge University Press, Cambridge, 1997).
[6] S. V. Dordevic *et al.*, Phys. Rev. Lett. **86**, 684 (2001).
[7] L. Degiorgi *et al.*, Z. Phys. B **102**, 367 (1997).
[8] C. Geibel *et al.*, Z. Phys. B **84**, 1 (1991).
[9] R. Caspary *et al.*, Phys. Rev. Lett. **71**, 2146 (1993).
[10] R. Feyerherm *et al.*, Phys. Rev. Lett. **73**, 1849 (1994).
[11] K. Knöpfle *et al.*, J. Phys. Condens. Matter **8**, 901 (1996).
[12] A. Krimmel *et al.*, J. Phys. Condens. Matter **8**, 1677 (1996).
[13] M. Jourdan *et al.*, Nature (London) **398**, 47 (1999).
[14] N. K. Sato *et al.*, Nature (London) **410**, 340 (2001).
[15] M. Huth *et al.*, Solid State Commun. **87**, 1133 (1993).
[16] M. Dressel *et al.*, cond-mat/0203308.
[17] A. A. Abrikosov *et al.*, *Quantum Field Theoretical Methods in Statistical Physics* (Pergamon, New York, 1965).
[18] The discrepancy between the high cutoff frequency compared to the superconducting gap at 4 cm^{-1} has implications on the kinetic energy and is discussed in [16].
[19] H. R. Ott, in *Progress in Low Temperature Physics,* edited by D. F. Brewer (Elsevier, Amsterdam, 1987), Vol. 11, p. 215.
[20] G. Zwicknagl *et al.*, Phys. Rev. B **65**, 081103 (2002).
[21] P. E. Sulewski *et al.*, Phys. Rev. B **38**, 5338 (1988).
[22] S. Donovan *et al.*, Phys. Rev. Lett. **79**, 1401 (1997).
[23] T. Timusk and B. Statt, Rep. Prog. Phys. **62**, 61 (1999).