Hybridization Gap in Heavy Fermion Compounds

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We report the results of optical studies of new heavy fermion compounds $YbFe_4Sb_{12}$ and $CeRu_4Sb_{12}$. We show that these compounds, as well as several other heavy fermion materials with a nonmagnetic ground state, obey a universal scaling relationship between the quasiparticle effective mass m^* and the magnitude of the energy gap Δ in the excitation spectrum. This result is in accord with the picture of hybridization of localized f-electron and free carrier states.

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Intermetallic compounds containing elements with f-electrons show a rich variety of effects, including a large enhancement of the quasiparticle effective mass [so-called "heavy fermion" (HF) behavior], a Kondo insulating state, and unconventional superconductivity [1–3]. These effects have been at the focus of condensed matter physics over the last two decades. The qualitative picture of both HF and the Kondo insulating state is based on the idea that ground state results from a competition between Kondo and RKKY interactions [4]. If the RKKY interaction dominates, various magnetic ground states can occur. However, if the Kondo interaction dominates, theory predicts that hybridization between localized f-electron and conducting carrier states should lead to the opening of a charge gap (or pseudogap) at the Fermi energy [5–9]. While this picture is not in dispute, the well-defined predictions of the hybridization scenario have so far escaped *direct* experimental verification. One of the key predictions is a simple scaling relationship between the magnitude of the direct energy gap Δ in the excitation spectrum and the enhancement of the effective mass of conducting carriers m^* in the coherent regime [10]. Several HF materials, such as $CeAl_3$ [11], show no evidence of such a gap, whereas in prototypal compounds such as UPt_3 and URu_2Si_2 the gap is attributed to a magnetic ground state [12–15].

In this paper, we present results for two new HF materials that belong to the filled skutterudite family, $YbFe_4Sb_{12}$ and $CeRu_4Sb_{12}$ [16–18]. In our view, $YbFe_4Sb_{12}$ and $CeRu_4Sb_{12}$ are perfectly suited to test the hybridization gap paradigm because the characteristic temperature T^* of the crossover to the coherent regime in the electronic transport in both compounds is as high as 50 K, permitting one to examine transport and optical properties at $T \ll T^*$. Based on magnetic susceptibility and electrical resistivity measurements [17,20], $CeRu_4Sb_{12}$ is speculated to order magnetically below 10 K, but in this work we studied only its paramagnetic phase.

Infrared spectroscopy is an ideal method to explore the heavy fermion ground state. This technique enables one to measure directly the magnitude of the gap in the density of states, while the analysis of the optical constants allows evaluation of the mass enhancement m^* over the free electron mass m_0 or the carrier band mass m_b [1,2]. The complex conductivity $\sigma(\omega) = \sigma_1(\omega) + i\sigma_2(\omega)$ was determined through Kramers-Kronig analysis from the reflectivity measurements carried out over the frequency range from 40 to 30 000 cm^{-1}. As a high energy extrapolation we used the data for the metallic skutterudite compound $LaFe_4P_{12}$, measured up to 12 eV [21]. As a low-frequency extension, we used the Hagen-Rubens formula and the dc resistivity values obtained for samples from the same batch.

Insight into the changes of the electronic structure and the carrier dynamics of $YbFe_4Sb_{12}$ and $CeRu_4Sb_{12}$ associated with the crossover to coherent transport is provided by the spectra of the dissipative part of the optical conductivity $\sigma_1(\omega)$ (Figs. 1 and 2). At $T > T^*$, the frequency dependence of the conductivity can be described with a simple Drude formula: $\sigma_1(\omega) = \sigma_0/(1 + \omega^2\tau^2)$, where σ_0 is the dc conductivity and τ is the carrier relaxation time. Besides this free electron contribution, in both compounds we observe an interband feature at $\sim$10 000 cm^{-1}, typical for a variety of skutterudites [21]. In addition, sharp peaks due to infrared active phonons were seen at 114 and 267 cm^{-1} for $YbFe_4Sb_{12}$ and at 116, 221, and 248 cm^{-1} for $CeRu_4Sb_{12}$. At $\omega < 90$ cm^{-1} for $YbFe_4Sb_{12}$, and $\omega < 400$ cm^{-1} for $CeRu_4Sb_{12}$, the low-T conductivity is suppressed compared to the spectrum at $T \approx T^*$ and shows a gap-like threshold at Δ (Figs. 1 and 2). The location of the gap feature is the same at all $T < T^*$, whereas the amount of the spectral weight $N_{\text{eff}}(\omega) = \int_0^\omega d\omega' \, \sigma_1(\omega')$ in the intragap region is strongly T dependent. The spectral weight from the intragap region is transferred primarily to higher energies and is completely recovered for $\omega < 0.2$ eV for $YbFe_4Sb_{12}$ and 0.6 eV for $CeRu_4Sb_{12}$. Interestingly, the energy scale involved in the redistribution of the spectral weight exceeds both Δ and T^* by at least a factor of 10. Such a mismatch

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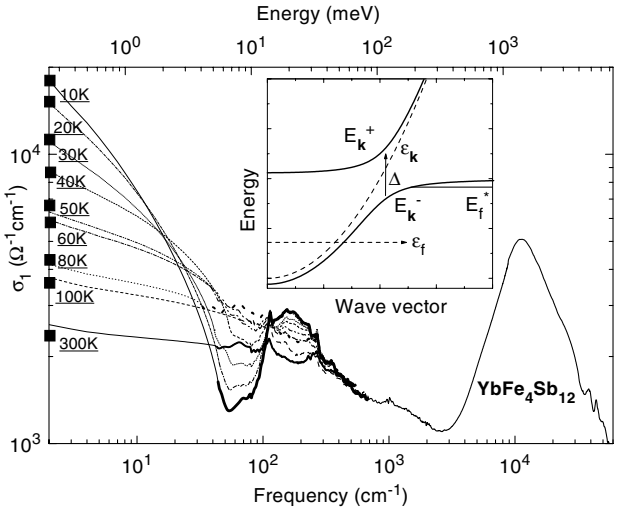

FIG. 1. Real part of the conductivity of $YbFe_4Sb_{12}$. The symbols on the left axis represent dc values at different temperatures. The thin lines are used in the region where reflectance was extrapolated with the Hagen-Rubens formula. Below T^*, a narrow peak at zero frequency and a gap-like feature at $\simeq$18 meV gradually develop. Inset: Renormalized band structure calculated from the Anderson lattice Hamiltonian. $\epsilon_{\mathbf{k}}$ and ϵ_f denote bands of free carriers and localized f-electrons. At low temperatures, a direct gap Δ opens. The Fermi level E_F^* is near the top of the lower band $E_{\mathbf{k}}^-$, resulting in hole-like character and enhanced effective mass of the quasiparticles.

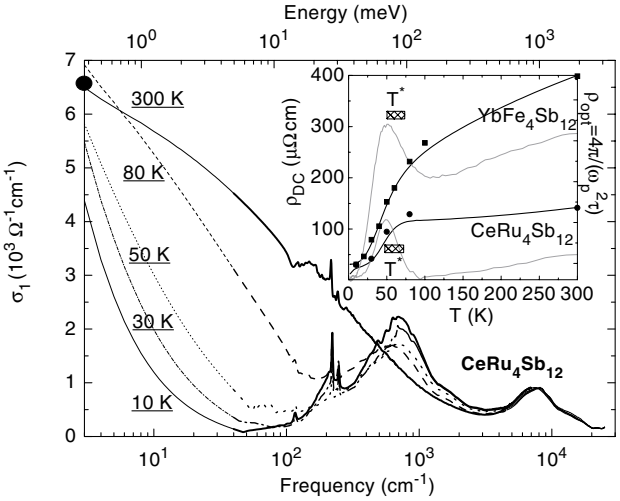

FIG. 2. Real part of the conductivity of $CeRu_4Sb_{12}$. Below T^* a very narrow peak at zero frequency and a resonance with an onset at 55 meV gradually develop. Inset: temperature dependence of the dc resistivity (left axis) and optical resistivity (right axis). The gray lines are the first derivative of the resistivity. T^* can be chosen either as the point where the resistivity suddenly drops or as the inflection point, i.e., the peak in the derivative. This leads to about 15% uncertainty in T^*.

has been previously detected in both metals and insulators with hybridization gaps in the density of states [22,23]. It is noteworthy that in the Yb-based compound, the gap affects only a small part of the Fermi surface, whereas the gap-free regions are responsible for a metallic background in the spectra of $\sigma_1(\omega)$ which is only weakly T dependent. In the Ce-based compound, the background is very small, and therefore the gap is likely to affect most of the Fermi surface.

Another distinct feature of the optical conductivity of both $YbFe_4Sb_{12}$ and $CeRu_4Sb_{12}$ in the coherent regime is a narrow resonance at $\omega = 0$. At $T > T^*$, we find good agreement between $\sigma_1(\omega)$ at the lowest measured frequency (40 cm^{-1}) and the dc value. Below T^*, such an agreement is no longer observed and dc values systematically exceed $\sigma_1(\omega = 40\ cm^{-1})$. This behavior is consistent with the development of a narrow (Drude-like) mode in the response of our samples. Such narrow modes, with half-widths of only a few wave numbers, are also suggested by the frequency dependence of the conductivity in the extrapolated region (thin lines in Figs. 1 and 2). Similar and even narrower resonances are found in the low-temperature conductivity of all HF metals [1,2]. Usually, these features are attributed to the renormalization of the quasiparticle scattering rate $1/\tau(\omega)$ and the effective mass $m^*(\omega)$. The extended Drude formalism is the canonical way to quantify renormalization:

$$\frac{m^*(\omega)}{m_b} = \frac{\omega_p^2}{4\pi}\,\frac{\sigma_2(\omega)}{\sigma_1^2(\omega) + \sigma_2^2(\omega)}\,\frac{1}{\omega}\,, \tag{1}$$

$$\frac{1}{\tau(\omega)} = \frac{\omega_p^2}{4\pi}\,\frac{\sigma_1(\omega)}{\sigma_1^2(\omega) + \sigma_2^2(\omega)}\,, \tag{2}$$

where the plasma frequency $\omega_p^2 = 4\pi e^2 n/m_b$ is estimated from the integration of $\sigma_1(\omega)$ up to the frequency of the onset of interband absorption. The spectra of the effective mass and of the scattering rate calculated from Eqs. (1) and (2) are plotted in Fig. 3. At $T > T^*$, the spectra of $1/\tau(\omega)$ are nearly frequency independent. As temperature is lowered below T^*, the absolute value of $1/\tau(\omega)$ in the intragap region is suppressed and a threshold feature emerges in the spectrum. Notably, the "optical resistivity", $\rho_{\rm opt} = 4\pi/\omega_p\tau^2(\omega \to 0)$, calculated from the data at the lowest measured frequency, reveals excellent agreement with the direct dc measurements. Suppression of the scattering rate is in accord with the strong enhancement of the effective mass in $YbFe_4Sb_{12}$ and $CeRu_4Sb_{12}$. We observe $m^*/m_b \sim 25$ and 80 for the Yb- and Ce-based compounds, respectively. These results are consistent with the specific heat measurements which yield $m^*/m_0 \approx 27$ and 90 [16]. The conclusion of electronic mass enhancement in $YbFe_4Sb_{12}$ and $CeRu_4Sb_{12}$ is robust and was tested using other approaches towards the analysis of the optical constants, i.e., "sum rule arguments" [24] and the ratio of zero crossings (Fig. 3) in the dielectric function ϵ_1 [1].

It is generally accepted that the essential physics of the HF metals and Kondo insulators is captured in the

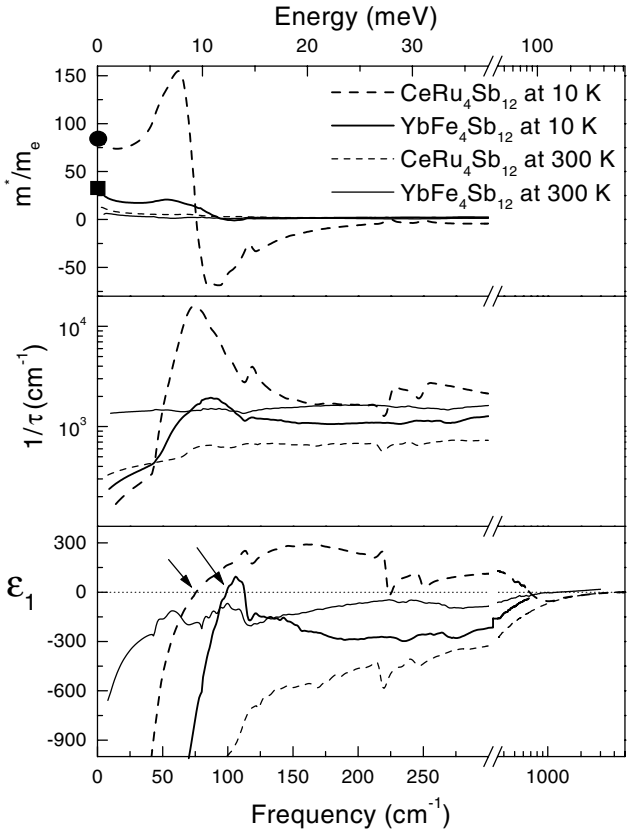

FIG. 3. Frequency dependence of the quasiparticle effective mass and scattering rate (top and middle panel, respectively) and real part of the dielectric constant ϵ_1 (bottom panel) for both $YbFe_4Sb_{12}$ and $CeRu_4Sb_{12}$. The symbols on the left indicate the effective masses obtained from specific heat measurements. The arrows point to low-frequency plasmons, which are due to heavy quasiparticles.

Anderson lattice Hamiltonian [1,6,9,25], describing a band of conduction electrons which hybridizes with localized f-electrons. Calculations show that for $T < T^*$, hybridization leads to a dispersion relation schematically shown in the inset in Fig. 1 [5,7,8]. This band structure gives excellent account of the behavior of $YbFe_4Sb_{12}$ and $CeRu_4Sb_{12}$. The hole-like character of conducting carriers, as inferred from Hall effect measurements [20], suggests that the Fermi level is in the lower band $E_{\mathbf{k}}^-$. At the Fermi energy the band is flat, in agreement with the renormalization of the effective mass and the scattering rate at low T (Fig. 3). The existence of heavy quasiparticles is confirmed by the observation of a low frequency plasmon in the spectra of the real part of the dielectric constant $\epsilon_1(\omega)$, Fig. 3. The hybridization picture also implies a gap in the density of states, giving rise to a second absorption channel in the conductivity spectra. A low-lying gap is found in both compounds (Figs. 1 and 2). Several numerical calculations of the optical conductivity exist [26,27] that are in qualitative agreement with our data. To determine the gap value accurately, we have performed a simple simulation of $[\sigma_{\mathrm{coh}}/\sigma_{\mathrm{incoh}}]$, the ratio of the optical conductivities in the coherent and incoherent states. The method follows closely the BCS calculation [28], except that the coherence factors are of type I, as opposed to type II, in conventional superconductors [29]. The low-temperature conductivity is obtained as $\sigma(\omega, 10\ \mathrm{K}) = [\sigma_{\mathrm{coh}}/\sigma_{\mathrm{incoh}}] \cdot \sigma(\omega, 50\ \mathrm{K})$. In the insets in Fig. 4, we show the final results of the simulation, indicated by gray lines. The excellent agreement allows very accurate determination of the magnitude of the gap.

From the band structure shown in the inset in Fig. 1, it is clear that there must be a relation between quasiparticle effective mass ($\frac{1}{m^*} \sim \frac{d^2E}{dk^2}$) and the magnitude of the gap. Indeed, calculations yield [5]

$$\frac{m^*}{m_b} \simeq \left(\frac{\Delta}{T^*}\right)^2. \tag{3}$$

As can be seen from Fig. 4, this scaling is in good agreement with our experimental data. The relation described by Eq. (3) is general and should hold for all paramagnetic HF materials. Indeed, we were able to identify similar scaling in at least two other paramagnetic compounds: $CeCu_6$ [30] and $CeCu_5$ [31–33]. If the correlation were to hold for another nonmagnetic HF compound $CeAl_3$, then the magnitude of the gap Δ can be expected to be as wide as 500 cm^{-1}. However, in the latter system the gap has not been observed [11], indicating that the quasiparticle concept may break down at such high energies.

It is noteworthy that HF materials with a magnetic ground state show systematic deviations from the scaling implied by Eq. (3), falling both above ($CePd_3$ [34]) and below (UPt_3 [14], URu_2Si_2 [13], U_2PtC_2 [2], and UCu_5 [12]) the universal line [35]. It is likely that magnetic

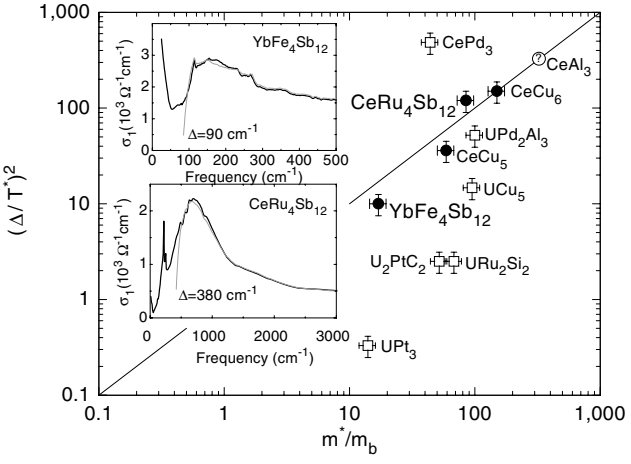

FIG. 4. A universal relation is predicted [5] between the effective mass of the quasiparticles m^* and magnitude of the gap Δ in the density of states of nonmagnetic HF compounds. HF compounds which order magnetically are not expected to satisfy the scaling relation and, in general, do not fall on the curve. The insets show the results of the BCS-like calculation of the optical conductivity, which allow accurate determination of Δ.

excitations further enhance the effective mass compared to the value derived from the hybridization mechanism. Alternatively, one can suggest that magnetic ordering may lead to the suppression of the energy gap. Both scenarios are consistent with the notion that in magnetic HF compounds the RKKY interaction overwhelms the Kondo coupling which is responsible for Eq. (3) [4]. One way to distinguish between the role of the two competing mechanisms in the formation of the heavy quasiparticle ground state is to explore the response of the HF compounds in which magnetic ordering at $T = T_M$ emerges out of the coherent state at $T < T^*$. At least two materials from Fig. 4 belong to this class: $CeRu_4Sb_{12}$ ($T_M \simeq 7$ K, $T^* \simeq 60$ K) and $CeCu_5$ ($T_M \simeq 4$ K, $T^* \simeq 6$ K); low-T experimental data are not available at the moment.

In conclusion, infrared experiments have revealed a scaling relationship between the magnitude of the effective mass m^* and the gap Δ that is followed by most nonmagnetic HF systems. In contrast, HF compounds which order magnetically show distinct deviations from this pattern, signaling the possibility of more than one single route to the heavy fermion state [36].

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