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# Possible non-Fermi-liquid nature of paramagnetic $UBe_{13}$: The optical point of view

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Optical investigations over a broad spectral range, extending from the far infrared up to the ultraviolet, reveal a peculiar and anomalous behavior of the excitation spectrum in $UBe_{13}$. With decreasing temperature down to 1.8 K a suppression of the Drude weight and a mid-infrared absorption are observed, similar to the optical properties of the so-called Kondo alloys with supposed non-Fermi-liquid behavior at low temperatures. A discussion of our experimental findings with respect to recent theoretical calculations, suggesting the non-Fermi-liquid nature of $UBe_{13}$, is proposed. [S0163-1829(97)52340-X]

Heavy-electron (HE) materials, which were discovered in the late 1970s,[1,2] have attracted a lot of interest because of their peculiar low-temperature properties distinguishing them from ordinary metals. The physics of these interesting anomalous metals is related to strongly correlated electrons in $4f/5f$ orbitals. Some of these systems are well described as Landau Fermi liquids (FL) of massive quasiparticles.[1,2] One of the most relevant questions in the field of highly correlated electron systems concerns the degree to which the phenomena are universal.

In fact, a number of so-called non-Fermi-liquid (NFL) HE alloys have been recently discovered, which display, e.g., a diverging linear coefficient of the specific heat $C(T)$, a strong $T$ dependence of the magnetic susceptibility, and a linear $T$ dependence of the resistivity for $T \rightarrow 0$.[3–5] Deviations from FL predictions have now been observed in several $f$-electron alloys, including $UCu_{5-x}Pd_x$ ($x=1$ and 1.5), $CeCu_{5.9}Au_{0.1}$, $Y_{1-x}U_xPd_3$ ($x<0.2$), and $U_{1-x}Th_xPd_2Al_3$ ($x>0.4$).[3–5] Besides these alloys, the superconducting $UBe_{13}$ also has NFL behavior in the specific heat and possesses a very large residual resistivity at the superconducting transition even in high-quality samples.[1,2] $UBe_{13}$ is, moreover, of particular interest since it is a concentrated ordered system, where the NFL state is not the consequence of alloying or of variation of some external parameters (magnetic field, pressure, etc.).

Anomalous screening of the magnetic impurity or vicinity to a magnetic instability are two possible scenarios for the development of NFL behavior.[3–5] Alternatively, and essentially for more dilute systems (like, e.g., $UCu_{5-x}Pd_x$), Miranda and co-workers suggested that the NFL behavior could be driven by disorder.[6] Cox proposed, based on symmetry grounds, that the NFL behavior of $UBe_{13}$ can be encountered by the two-channel Kondo model description.[7] In this picture, electrical quadrupole moments of the twofold nonmagnetic $\Gamma_3$ ground state of the U ion are screened by orbital motion of the conduction electrons, which supply two screening channels.[7] These conduction channels couple ‘‘antiferromagnetically’’ to the local quadrupole ‘‘spin,’’ leading to a so-called overcompensated quenched impurity state with ‘‘marginal’’ Fermi-liquid behavior in the transport, magnetic, and thermodynamic properties.[7] More recently, Anders, Jarrell, and Cox tackled the problem of the corresponding lattice model.[8] They also performed a calculation

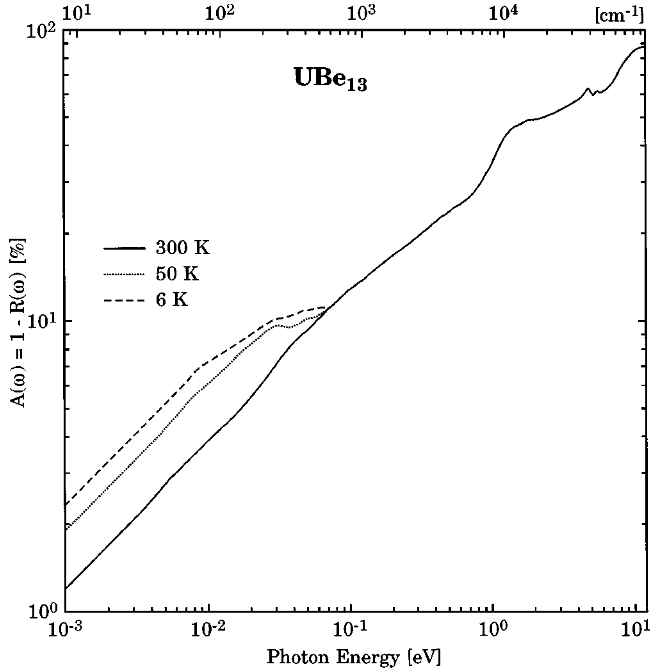

FIG. 1. Frequency dependence of the absorptivity of $UBe_{13}$ at several relevant temperatures.

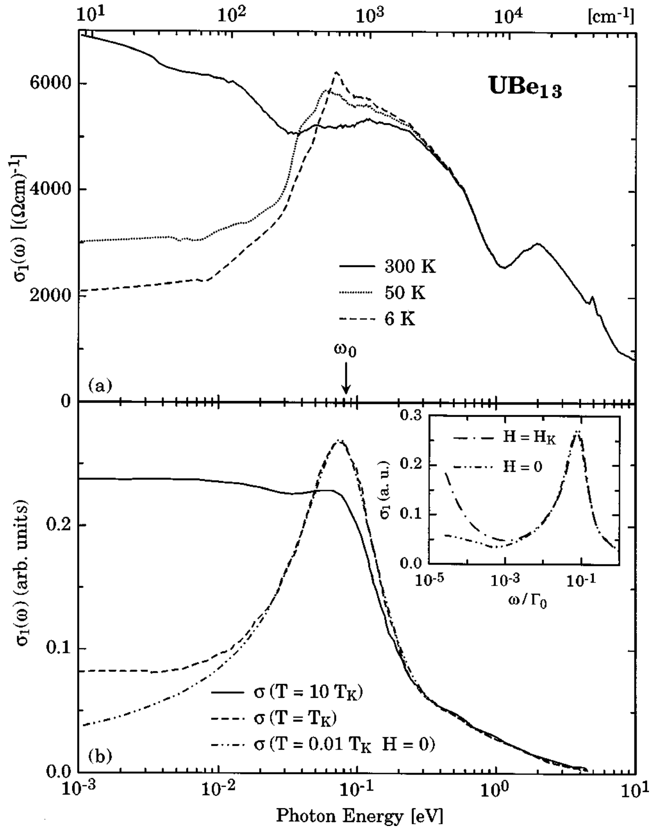

FIG. 2. (a) Real part $\sigma_1(\omega)$ of the optical conductivity of $UBe_{13}$ at 300, 50, and 6 K. (b) The calculated $\sigma_1(\omega)$ at $T=10T_K$, $T_K$, and $0.01T_K$ in zero applied magnetic field within the two-channel Anderson lattice model (Ref. 8). One should note the suppression of the Drude peak and the corresponding mid-infrared absorption at $T\leqslant T_K$ and the reappearance of the Drude contribution at $T=0.01T_K$ by applying the magnetic field $H_K$ (inset).

of the optical properties within such a two-channel Anderson lattice model for which the suppression of the low-frequency Drude component and the development of a mid-infrared absorption in the excitation spectrum at low temperatures have been suggested.[8]

Previous investigations on typical Kondo alloys pointed out that the mapping of the complete electrodynamic response leads to frequency and temperature dependencies of the scattering relaxation rate $\Gamma$ which deviate remarkably from the FL prediction (i.e., $\Gamma\sim\omega^2+T^2$).[9–11] We were then motivated to apply optical spectroscopic tools to $UBe_{13}$ in order to study its excitation spectrum and the possible implications with respect to its supposed NFL nature. This paper presents our first efforts to address this issue. Our optical data display the disappearance in $\sigma_1(\omega)$ of the Drude weight at low frequencies and temperatures in favor of a mid-infrared absorption, in good agreement with the theoretical predictions.[8]

The optical reflectivity on a $UBe_{13}$ single crystal, grown as described in Ref. 12, has been measured over a broad spectral range extending from the far-infrared (FIR) up to the ultraviolet.[9–11] The optical conductivity is obtained from Kramers-Kronig (KK) transformations applied to the measured optical reflectivity. Appropriate extrapolations were used above the highest frequency limit, while below the lowest measured frequency we performed an *ad hoc* Hagen-Rubens extrapolation to frequency zero (see below).[1,2]

Figure 1 displays the optical absorptivity $A(\omega)=1-R(\omega)$ at some relevant temperatures, while Fig. 2(a) shows the corresponding real part $\sigma_1(\omega)$ of the complex optical conductivity. The logarithmic energy scale of both figures allows the presentation of the whole excitation spectrum but obviously emphasizes the temperature dependence of both quantities in FIR. The absorptivity is characterized by a typical plasma edge feature at about 10 eV, where $A(\omega)$ has its typical onset for a metallic behavior [i.e., $R(\omega)=1-A(\omega)$ increases continuously so that $R(\omega)\rightarrow 100\%$ for $\omega\rightarrow 0$]. Overlapped to the plasma edge feature and at higher frequencies there are several electronic interband transitions. Our results, which to our best knowledge are obtained for the first time over a very broad spectral range on a single crystalline specimen, are in fair agreement with previous optical data, obtained on limited spectral ranges by different groups.[13–15] Particularly, Bonn *et al.* suggested an anomalous or unconventional metallic behavior of $\sigma_1(\omega)$ at low temperature.[15]

From now on we will mainly focus on the FIR optical properties. The most striking feature is, indeed, the increasing $A(\omega)$ (Fig. 1) in FIR at low temperatures, which leads to the suppression of the FIR spectral weight in $\sigma_1(\omega)$ [Fig. 2(a)]. Alternatively, one can say that the Drude peak is progressively suppressed with decreasing temperature, leading to a mid-infrared feature (i.e., the absorption at about 0.1 eV) in $\sigma_1(\omega)$. It is worth noting that our measurements between 6 and 1.8 K did not show any further temperature dependence.

The dc limit $\sigma_1(\omega\rightarrow 0)$ is in fair agreement with the $\sigma_{dc}$ values obtained from transport properties' investigations on different crystals.[1,2] Such an agreement is well realized at 300 K, while at temperatures lower than 100 K there is a systematic deviation with $\sigma_1(\omega \rightarrow 0) \leqslant \sigma_{dc}$, but still indicative of the high quality of the sample. Surface scattering effects, which might be of relevance in highly conducting materials even in the FIR range (i.e., at low temperatures the larger $\sigma_{dc}$ is, the smaller the penetration depth, and the more important the shape of the surface), can possibly explain the deviations between $\sigma_1(\omega \rightarrow 0)$ and $\sigma_{dc}$.

Moreover, one can use the surface resistance ($R_S$) measurement of $UBe_{13}$ at 102 and 148 GHz obtained by Beyermann and co-workers.[16] The direct calculation of $\sigma_1(\omega) \sim \omega/R_S^2$ from the $R_S$ measurements gives reasonable agreement, even though the implicit Hagen-Rubens limit (i.e., $R_S = X_S$, since the surface reactance $X_S$ was not measured) might be too crude.[16] This latter approximation is neither affecting the main trend of $\sigma_1(\omega)$ envisaged by our KK results nor the content of the following discussion. An alternative way to integrate our optical data with the microwave points[16] (102 and 148 GHz) could be to regain first the absorptivity at those frequencies [i.e., $A(\omega) = 4R_S/Z_0$, $Z_0$ is the free space impedance], and to combine it with the FIR absorptivity of Fig. 1. One can perform the Kramers-Kronig transformation once again, in order to get $\sigma_1(\omega)$. In this way the approximation $R_S = X_S$ is less damaging since for highly conducting samples the condition $R_S, X_S \ll Z_0$ is always realized, leading to $A(\omega)$ depending from $R_S$ only.[16] Even in this way, the general shape of $\sigma_1(\omega)$ with decreasing temperature is confirmed, as displayed in Fig. 2(a). Such a behavior of $\sigma_1(\omega)$ for $UBe_{13}$ bears a remarkable similarity with optical results recently obtained on so-called NFL Kondo alloys, as $U_{0.2}Y_{0.8}Pd_3$,[9] $U_{1-x}Th_xPd_2Al_3$ $(x > 0.4)$,[10] and $UCu_{5-x}Pd_x$ $(x = 1,1.5)$.[11] Putting this observation in the broader context of the electrodynamic response of correlated metals, it is worth noting that for typical HE materials, as $CeAl_3$ or $UPt_3$, $\sigma_1(\omega)$ displays on the contrary an enhanced behavior of $\sigma_1(\omega)$ at low frequencies and temperatures[17,18] instead of the spectral weight suppression. Such a narrow Drude-like behavior is ascribed to the heavy quasiparticles' contribution to $\sigma_1(\omega)$.

As briefly anticipated in the introduction, the calculation of $\sigma_1(\omega)$ by Anders, Jarrell, and Cox[8] within the two-channel Anderson lattice is very compatible with the experimental data presented here for $UBe_{13}$ and elsewhere for a series of Kondo alloys.[9–11] Theoretically, $\sigma_1(\omega)$ at low temperatures displays a large charge fluctuation peak at an energy scale of approximately $\omega_0 \sim 10T_K$, $T_K$ being the Kondo temperature [see Fig. 2(b)]. From a first comparison between optical data and theory we have estimated $T_K \approx 10$ K. Furthermore, the removed spectral weight piles up at the peak $\omega_0$, in accordance with the $f$-sum rule.[19,20] This latter model does not take into account scattering due to phonons or high-energy electronic interband transitions. Obviously, these scattering processes can considerably affect the absolute magnitude of the optical conductivity and explain the decrease of $\sigma_1(\omega)$ above 0.2 eV in the theoretical curve [Fig. 2(b)]. Therefore, the comparison can be performed on a qualitative level only. The agreement with the available experimental data is outstanding and all major features predicted theoretically are encountered in our experimental data (see Fig. 2). Finally, it is worth mentioning that the calculated $\sigma_1(\omega)$ does not display any clear Drude peak even for temperatures as low as $T = 0.01T_K$, one decade lower than the observed maximum in $\rho(T)$.[1,2,8] In fact down to 1.8 K, we do not recover any sign of the Drude contribution in $\sigma_1(\omega)$.

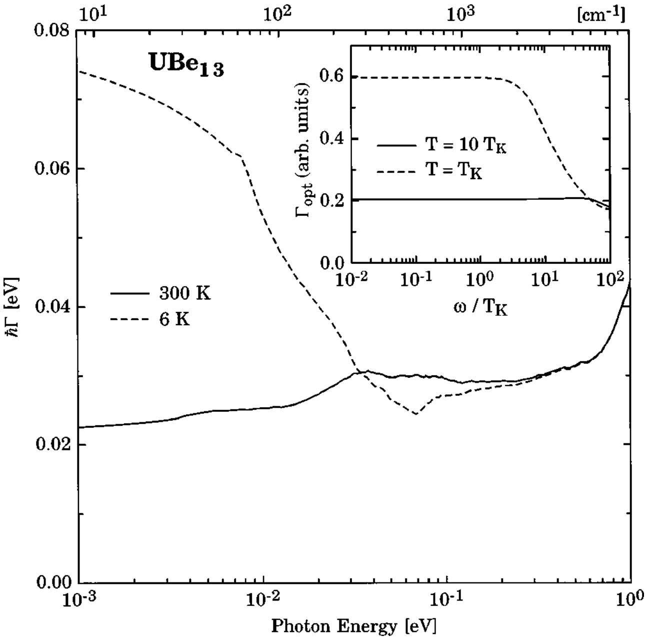

FIG. 3. Frequency dependence of the scattering rate (Γ) at 300 and 6 K. The inset shows the theoretical prediction after Ref. 8.

Otherwise, one might argue that the suppression of Drude weight at low frequencies is nothing else than the natural and direct consequence of the large dc resistivity, leading to a depressed conductivity (compared to the higher conductivity which occurs at higher temperatures), and of the spectral weight shift, owing to the conservation of the spectral sum rule. However, the striking contrast between the excitation spectra of $UBe_{13}$ and those of other ordered highly correlated metals, as $CeAl_3$ and $UPt_3$, is certainly peculiar. The two-channel Anderson lattice model with its implicit marginality provides a possible microscopic mechanism. Nonetheless, it is well known that $UBe_{13}$ is clearly not in its ground state at the temperatures where the measurements were made and the application of the model of Ref. 8 is only valid in the paramagnetic phase. Furthermore, the vicinity of the superconducting phase transition could lead to some kind of singularity or fluctuation effects.

One can further exploit the optical data by extracting the temperature and frequency dependence of the scattering relaxation rate $\Gamma$. This can be achieved by applying a so-called generalized Drude model.[9–11] This method is particularly compelling at low frequencies, where no obvious method exists for separating $\sigma_1(\omega)$ into an ordinary Drude contribution and mid-infrared modes. In general, the generalized Drude model allows us to reveal a fundamental relationship between the scattering rates derived from either dc transport properties or the optical response in FIR. The frequency dependence of the scattering rate at some relevant temperatures is shown in Fig. 3. For the two-channel Anderson lattice model,[8] the optical scattering rate is nearly frequency independent at low frequencies in fair agreement with the experimental estimation of $\Gamma_{opt}$, which tends to saturate towards a constant value (see Fig. 3 and its inset).[8] However, one should keep in mind that $\Gamma_{\rm opt}$ reflects the two-particle nature of the energy absorption process associated with electrical charge transport. Generally, only for a Fermi liquid at very low temperatures and frequencies should $\Gamma_{\rm opt}$ coincide with the one-particle relaxation rate.[21] Therefore, the proposed NFL nature of $\mathrm{UBe}_{13}$ might partially invalidate such a comparison based on the scattering rate. It is, however, interesting to notice that the correspondence between theory and experiment in terms of the scattering rate (more precisely through the evaluation of the so-called complex self-energy) seems to be a common feature of NFL Kondo alloys, as well.[9–11] It remains to be seen how far one can push such an argument, since the quasiparticle concept somehow collapses in NFL systems.

In summary, the complete electrodynamic response of $\mathrm{UBe}_{13}$ obtained over several decades in frequencies displays a rather peculiar behavior at low temperatures. This is evidenced most clearly by the suppression of the Drude component in FIR. Such a behavior is in agreement with the major predictions of the model applied by Anders, Jarrell, and Cox[8] and could imply a NFL behavior. However, other approaches can lead to similar results: as the quite similar two-channel Kondo lattice[22,23] and the disordered-driven NFL behavior.[6,24] It is an essential and relevant task to find a way where one can discriminate between different models by selectively changing or tuning some intrinsic parameters (i.e., either by alloying or by varying external parameters like pressure and magnetic field). In this respect and particularly as far as $\mathrm{UBe}_{13}$ is concerned, the calculation by Anders, Jarrell, and Cox predicts that a low-frequency ‘‘Drude’’ peak, similarly to the situation encountered in $\mathrm{CeAl}_3$ or $\mathrm{UPt}_3$,[17,18] should develop again at low temperatures in a magnetic field of $H=H_K$, consistent with the return to FL behavior [see inset Fig. 2(b)].[8] This is an interesting prediction that can be used to test the model or alternatively to single out the most appropriate theoretical approach. Consequently, low-temperature optical experiments on $\mathrm{UBe}_{13}$ in a magnetic field and possibly extended to even lower frequency (i.e., below the FIR spectral range) are clearly desirable.

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