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Rapid Note

Charge excitations in heavy electron metals

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Abstract. We show that the optical response of metals with strong electron-electron correlation consists of two excitations, a renormalized Drude response at zero energy and a mid-infrared peak occurring at frequencies around 2000 cm^{-1}. The latter originates from a dynamical, correlation-induced gap, as evinced from a many body theoretical approach based on the periodic Anderson model. At very low temperatures, it can be viewed as optical gap between two renormalized quasi-particle bands. The gap size is proportional to the geometric mean of the characteristic lattice Kondo temperature of the material and its bandwidth.

PACS. 78.20.-e Optical properties of bulk materials and thin films – 71.28.+d Narrow-band systems; intermediate-valence solids

The unusual thermodynamic, magnetic and transport properties of transition metals, where electron-electron correlations are strong, have been the subject of intense studies [1–4]. Much of the experimental findings can be described in terms of a coherent Fermi liquid (FL) state, which develops at low temperatures. Renormalization effects lead to enhanced thermodynamic quantities, such as specific heat, and these are usually described in terms of a large effective mass m^*, hence the name "Heavy Fermion" (HF). The renormalization is temperature dependent and governed by the characteristic temperature scale T^*, the lattice Kondo temperature. This crossover temperature measures the dynamical screening of the $4f(5f)$ moments through the conduction electrons. The temperature, where a smooth and gradual crossover to the heavy FL state is observed, is usually referred to as a coherence temperature $T_{\rm coh} < T^*$ [1–4].

The resistivity of these materials is large at room temperature T, usually increases with decreasing T to a maximum, which correlates with T^*, followed by a rapid drop which asymptotically exhibits a $\rho(T) = \rho_0 + AT^2$ behaviour for $T < T_{\rm coh}$ [1,2]. The scattering probability is strongly suppressed at $T < T^*$, due to the formation of a coherent state, whose quasi-particle excitations consist of a temperature and energy dependent mixture of conduction electrons and f-electrons. The observed unusual optical properties [5] reflect the many-body nature of the HF state: one observes a narrow resonance at zero frequency with a reduced spectral weight (reflecting a large electrodynamical mass m^*, see below) at low frequencies. This so-called renormalized Drude response has been extensively studied and the relation between the electrodynamics and the thermodynamics quantities is well-established [5].

In this communication we revisit and describe the overall optical response of a large variety of HF metals. Our goal is first to catch the essential experimental features, stressing their generality in the dynamics of HF, and second to suggest a comprehensive theoretical understanding. We show that in the coherent low temperature state there are two distinct charge excitations. We find a well-defined mid-infrared mode, together with the narrow renormalized Drude response identified earlier [5] in nearly all the materials in their HF state. There is a system-to-system variation but in most cases the mid-infrared resonance is in the spectral range between 1000 and 2000 cm^{-1}. Traditional band-structure calculations, based on a local density approximation (LDA), cannot explain these features, since the energy scales of interest are too small to be resolved, and the many-body nature of the HF is not accounted for. We will argue, that a many-body picture using renormalized quasi-particles [6] explains the occurrence and the location of the two peaks at zero and finite energy in terms of intra and inter-band transitions, respectively [7–10]. Additionally, this picture predicts a weak material dependence of the mid-infrared (MIR) peak, in agreement with our experimental findings.

We have performed optical reflectivity measurements as a function of temperature in an extremely broad spectral range, extending from the far infrared (FIR) up to the ultraviolet (UV). Our investigations also included frequencies below the conventional optical spectral range; namely, in the micro and mm wave range, where cavity resonant

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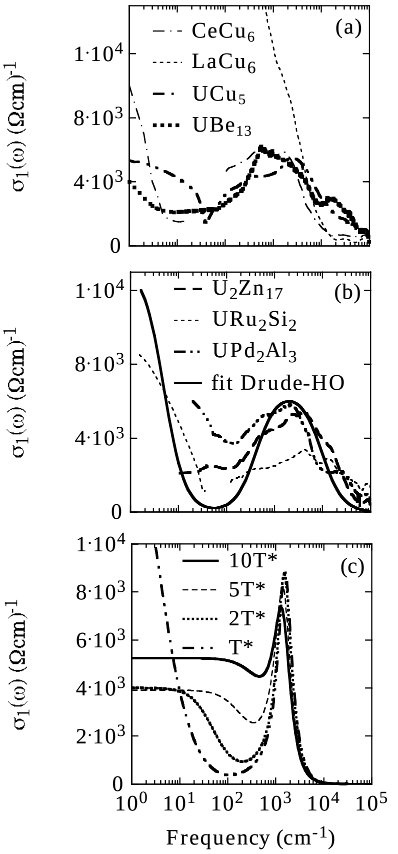

Fig. 1. (a)–(b) Frequency dependence of the real part $\sigma_1(\omega)$ of the optical conductivity for several heavy electron metals in their coherent low temperature phase [5] and, as comparison, for $LaCu_6$. The temperature was different from compound to compound but anyway lower than 10 K. As guide to the eyes and for a better highlight of the two components we display the Lorentz-Drude curve in part (b), as well. The ZE mode is calculated with a Drude plasma frequency of 2000 cm^{-1} and a scattering rate of 5 cm^{-1}; the Lorentz harmonic oscillator for the FE mode has a resonance frequency of 2000 cm^{-1}, a broadening of 10^4 cm^{-1} and a mode strength of 6×10^4 cm^{-1}. (c) Theoretical curves of $\sigma_1(\omega)$ with $T^* = 20$ K and $W = 2.3$ eV at $T = 1$, 2, 5 and $10T^*$, after equation (5) of reference [20].

methods have been applied [11,12]. Standard Kramers-Kronig transformations were employed in order to obtain the optical conductivity. We have measured several materials both in single crystalline as well as in polycrystalline form, with consistent results. Specific information about the crystal growth and technical details are summarized in a variety of publications (see Ref. [5] for a review). The optical results presented here are in broad agreement with previous and in some cases less detailed studies (where such results are available).

In Figure 1a–b we summarize the real part $\sigma_1(\omega)$ of the optical conductivity for several HF metals [5] at low temperatures where according to a variety of thermodynamic and transport measurements the coherent state is well developed. For the purpose of clarity, for some materials we have omitted from the total optical conductivity in Figure 1a–b the far-infrared excitations, associated with the various magnetic states developing in some HF materials [5]. For instance in URu_2Si_2 and UCu_5 a FIR absorption (*i.e.*, at energies below 100 cm^{-1}) was found at low temperatures and was ascribed to a spin density wave-like gap due to the incommensurate anti-ferromagnetic phase transitions while in UPt_3 a similar absorption at about 10 cm^{-1} was assigned to the onset of magnetic correlations [5]. These latter features and their origin are not addressed by the following theoretical approach.

A few features are evident: there is first of all a well-defined peak (FE) around 1000−2000 cm^{-1} with a maximum conductivity of about $6000\,(\Omega\,\mathrm{cm})^{-1}$ for most compounds. In order to further stress the peculiarity of such a mid-infrared absorption, we also show in Figure 1a the optical conductivity of $LaCu_6$, as comparison [13]. For $LaCu_6$, there is only a Drude-like metallic component [14]. Second, there is the narrow mode centered at zero frequency (ZE) which was recognized early and thoroughly discussed experimentally [5] and theoretically [8,9]. For comparison, the total spectral weight is $\omega_\mathrm{p}^2/8$ and is located mainly in the FE peak; ω_p gives the unrenormalized plasma frequency, which is of the order of a few electron volt. This plasma mode is clearly seen in the optical reflectivity as the high frequency plasma edge [5]. It is important to note that the finite energy mode basically contains the spectral weight associated with the unrenormalized band-mass m_B. The spectral weight of the ZE mode is significantly smaller than the total spectral weight and varies from material to material. It has been shown earlier that it is given by [5]:

$$\int_0^{\omega_\mathrm{c}} \mathrm{d}\omega \sigma_1(\omega) = \frac{\pi n e^2}{2m^*} = \frac{(\omega_\mathrm{p}^*)^2}{8} \tag{1}$$

with m^* the electro-dynamical effective mass at low frequency, which is enhanced by electron-electron interactions and $\omega_\mathrm{c} \approx 100\ \mathrm{cm}^{-1}$. This effective mass, obtained using the sum rule in equation (1), is in good agreement with the enhanced thermodynamic mass extracted from specific heat and magnetic susceptibility [1,2,5]. Therefore, we can conclude, that the dynamical mass-enhancement occurs only for energies of a few times $k_\mathrm{B}T^*$ around the chemical potential. In order to highlight the two components, we add to the figure (b) the phenomenological curve based on the Lorentz-Drude model [15].

It is believed that the periodic Anderson model (PAM) is the reasonable description of HF systems (for a review see Ref. [3]): localized $4f(5f)$ electrons hybridize locally with a free conduction electron band. Since localized electrons are dispersionless ($\epsilon_\mathrm{loc}(\mathbf{k}) = \text{const.}$), only the fraction of heavy quasi-particles, which have conduction electron character, contributes to the electrical current [16]. The optical conductivity is a response function, defined as:

$$\sigma(\omega) = \sigma_1(\omega) + \mathrm{i}\sigma_2(\omega) = n\frac{1}{\mathrm{i}\omega}\lim_{\mathbf{q}\to 0}\left(\langle j_\mathbf{q}|j_\mathbf{q}^\dagger\rangle(\omega) - S\right), \tag{2}$$

where n is the carrier concentration and

$$S = \frac{e^2}{\hbar^2}\frac{1}{N}\sum_\mathbf{k} \partial_k^2 \epsilon_\mathbf{k} \langle n_\mathbf{k}\rangle \,. \tag{3}$$

$\langle n_{\mathbf{k}}\rangle$ is the average number of electrons with momentum $\mathbf{k}$ and $\epsilon_{\mathbf{k}}$ is the unrenomalized conduction-electron band structure. This expression is derived from Gauge-invariance [17,18]. S is real and frequency-independent [19]. If anisotropy and the variation of the unrenormalized density of states $\rho(\omega)$ near the chemical potential μ are neglected, the optical conductivity is approximated by:

$$\frac{2\hbar^2}{ne^2a^2}\sigma(\nu) = \sum_{\sigma}\frac{\mathrm{i}W^2\pi\rho(\mu)}{\nu} \times \int_{-\infty}^{\infty}\frac{\mathrm{d}\omega}{2\pi}\,\frac{f(\omega)-f(\omega+\nu)}{\nu+\Sigma_{\mathrm{c}}(\omega-\mathrm{i}\delta)-\Sigma_{\mathrm{c}}(\omega+\nu+\mathrm{i}\delta)} \qquad (4)$$

where the volume of the unit cell is a^3, W is the band-width and $\Sigma_{\mathrm{c}}(\omega)$ the $\mathbf{k}$-independent self-energy of the conduction electrons [8,9]. Equation (4) is a useful approximation of equation (5) in reference [20], permitting a better identification of the relevant components in $\sigma_1(\omega)$. Any structure in the optical conductivity is caused by the dynamical renormalization of the band electrons. $\sigma_1(\omega)$ is governed by the imaginary part of the integral kernel in equation (4). The kernel becomes large, when

$$\nu \approx \Re e\Sigma_{\mathrm{c}}(\omega+\nu) - \Re e\Sigma_{\mathrm{c}}(\omega) \qquad (5)$$

for frequencies within the Fermi-range. This is obviously true at zero frequency, leading to the Drude-peak, which is caused by intra-band excitations around the chemical potential, and therefore is mainly influenced by the single particle relaxation rate $\Im m\Sigma_{\mathrm{c}}$. The new renormalized quasi-particles bands in PAM are determined by the solutions of the non-linear equation

$$\omega_{\lambda\mathbf{k}} - \epsilon_{\mathbf{k}} - \Re e\Sigma_{\mathrm{c}}(\omega_{\lambda\mathbf{k}}) = 0. \qquad (6)$$

Hence, equation (5) is also fulfilled, when the optical frequency ν matches the energy difference of two different solutions of equation (6) for the same momentum $\mathbf{k}$. This corresponds to an optical inter-band transition of hybridized bands close to the chemical potential. The band gap originates in the coherent resonant scattering of conduction-electrons on the Abrikosov-Suhl resonance of f-shells; it is pinned slightly above the chemical potential as a consequence of the Friedel-sum rule [21]. At temperature $T > T^*$, the quasi-particles picture is inadequate due to large life-time broadening. Although the resonance is broaden and becomes invisible for $T > T^*$, since spectral weight is removed from the spin into the charge-excitation, its location does not change, since the number of f-electrons is almost temperature independent.

The location of the inter-band peak is crudely estimated by calculating the optical gap, using a simple phenomenological approach for the hybridized bands [6]. Assuming a bandwidth of 2.3 eV and a characteristic $T^* \approx 20$ K, we obtain $\Delta_{\mathrm{opt}} \approx \sqrt{(8T^*W/\pi)} \approx 800\ \mathrm{cm}^{-1}$, which is lower than the calculated and measured value since the spectral weight above the chemical potential is underestimated. Nevertheless, it serves as a good estimate and backs the physical picture on which it is based. Experimentally, this implies that the wealth of possible combinations in T^* and W for a large variety of compounds would always cooperate in defining an approximated Δ_{opt} in rough agreement and consistent with the measured broad FE mode. A detailed discussion of the optical conductivity in a moderate correlation regime [22] can be found in reference [4]. Our calculations connect theory with experiment and demonstrate, that using only the experimentally observed T^* and carrier concentration in conjunction with a reasonable band width, we can reproduce the essential features of the experiment in terms of shape and magnitude of the optical conductivity spectrum over a broad range of frequencies without any fit-parameters. Previously, a similar formalism was mainly employed in order to reproduce the narrow Drude components only [3,6]. New numerical methods now available allow more reliable input in the optical conductivity [4,23,24].

In Figure 1c we show the calculated curve for the optical conductivity using equation (5) of reference [20] based on a simple single channel periodic Anderson model but taking into account various correction terms (*e.g.*, due to the non-constant $\rho(\omega)$) neglected in our (approximated) equation (5). Besides the values of T^* and W mentioned above we used $n = 0.8\ \mathrm{cm}^{-3}$ as a parameter. No additional impurity scattering has been taken into account. The valence fluctuations of the f-shells are restricted to two configurations: we consider the strong correlation limit. The conduction-electron self-energy is obtained by a dynamical mean field calculation [23]. The overall agreement with experiment is very good. Clearly visible is the ZE Drude peak that narrows on lowering the temperature (note the logarithmic scale). As similarly pointed out in reference [6] and [24], the FE peak can be seen at low temperatures as a manifestation of inter-band excitations across the hybridized quasi-particle bands. The effective hybridization strength is strongly reduced, since only f-electrons of the order of T^*/Δ participate in the resonant scattering [6].

The carrier density for the theoretical curve is adjusted to one electron per unit cell of size $a = 12$ Å, so that the magnitude of the low-frequency spectrum matches with the experimental findings. This yields a calculated plasma-frequency of 4.6 eV [25], while experimentally, we obtain 7 eV. The integrated spectral weight of the optical conductivity essentially counts the optically active degrees of freedom. It is not surprising, that the total spectral weight of the calculated optical conductivity is lower than the experimental one. Only the lowest Hund's rule crystal electric field state of the f-electrons and a single band with a Gaussian band density of states have been taken into account in the calculation. In the experiment, all ionic f-states contribute to the scattering of conduction electrons at higher frequencies due to quantum-fluctuation even in the ground-state. This leads to additional structure in the MIR and FIR region. Therefore, the width of theoretical MIR peak is smaller and has less structure than the experimental findings. Moreover, the crystal electric field induced additional structure and broadening of MIR peak reduce further the material dependency of the effective peak position whose location was estimated at Δ_{opt}. An artificial broadening of the theoretical curve could take into account impurity scattering and neglected quantum-fluctuations, and a larger carrier concentration could be assumed. However, no additional physical insight would be obtained, where and why the MIR peak occurs. Since T^* and W enter the optical gap only through a square root and the range of T^* is limited by the class of materials under investigation, Δ_{opt} does not strongly vary from compound to compound and give a very good estimate, where to expect the location of the mid-IR peak.

Concerning the temperature dependence, we notice that correlations affect the dynamics at any temperature. Even though the quasi-particle picture is not adequate any more at temperatures $T > T^*$, the single particle relaxation rate is only very weakly temperature-dependent at the MIR frequencies. This reflects the separation of high and low energy dynamics mentioned earlier in the paper and which is also seen in the experimental optical relaxation rate $\Gamma(\omega)$ [5]. Hence, the MIR peak persists up to high temperatures well above the lattice Kondo-temperature as observed [5]. The optical gap is strongly governed by local physics at $T > T^*$ and mainly singlet-triplet excitations occur on an energy scale of $k_{\text{B}}T^*$. Therefore, the MIR peak position is shifted sightly to lower frequencies compared to the coherent state as seen in Figure 1c and in the experimental data [5]. The imaginary part of the self-energy at frequencies $\omega \gg k_{\text{B}}T^*$ is also very weakly temperature-dependent, so the FE inter-band peak position is also expected to change very little with temperature. By lowering the temperature, correlation effects dressing the free charge carriers are manifested through a redistribution of spectral weight between higher and lower frequency as consequence of the enhancement of both the effective mass and the scattering time (*i.e.* narrowing of the Drude-like component in $\sigma_1(\omega)$).

Summarizing, the optical experimental results on HF point to common features, which are accounted for by the periodic Anderson model [3]. We have advanced a simple explanation of the MIR peak in terms of optical inter-band excitations. It is worth mentioning that recent angle-resolved photoemission spectroscopy studies [26] on f-electron materials are consistent with the physical picture presented here. The rather weak material dependence, found experimentally, of the peak position is in perfect agreement with the theory. While in the case of heavy electrons the heavy particle is a quasiparticle renormalized by electron-electron interactions, we believe that similar explanations could be advanced for other strongly correlated metals, as well. For instance, we believe that the recent optical measurement [27] on the mixed-valent state of YbInCu_4 can be understood along the same line. Moreover, it is worth mentioning in view of the generality of the experimental results that FE peaks in the MIR range are shared by variety of materials, such as the high temperature superconductors, the organic Bechgaard salts or the charge density wave systems [11,12,28,29], where electron-electron correlations or even electron-phonon interactions are important.

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