Signature of Kondo breakdown quantum criticality in optical conductivity

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We show that the behavior of the finite frequency inter-band transition peak in the optical conductivity of the heavy fermions can provide definitive experimental proof of the Kondo breakdown phenomenon in which the lattice Kondo temperature vanishes at a quantum critical point. Approaching such a phase transition from the heavy Fermi liquid side, we find a new cross-over regime where the peak position is related to, but is not directly proportional to the lattice Kondo scale. The peak position moves to lower energies but remains finite, while the peak value changes non-monotonically and it eventually disappears at the quantum critical point. These are unique signatures which distinguish a Kondo breakdown transition from a spin density wave driven transition.

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Introduction.— Developments in material synthesis have led to the discovery of numerous rare earth compounds, the heavy fermions (HFs), which can be tuned to a quantum critical point (QCP), separating a magnetic ground state from a paramagnetic one \cite{1}, by varying an external parameter such as pressure or chemical doping. In the quantum critical regime the metallic properties are significantly different from what one expects from a standard Landau Fermi liquid (LFL), and therefore these systems are prototypes to study how strong correlation effects give rise to deviations from LFL.

The early theoretical attempts to describe the QCP are based on the possibility that the instability of the paramagnetic phase is due to spin density wave (SDW) formation \cite{2}, and the critical fluctuations are the para-magnons. However, in three dimensions these theories fail to explain simultaneously the linear temperature \((T)\) dependence of the resistivity and the \(\log T\) dependence of the specific heat coefficient observed in experiments \cite{3}. This has stimulated theorists to construct alternative descriptions of the quantum criticality \cite{1}, among which the scenario of Kondo breakdown (KB) has proven to be promising \cite{3, 6}. In the KB scenario the non-LFL behaviour is due to the presence of a second QCP, in close proximity to the magnetic one, where the effective Kondo temperature of the lattice \((T_K)\) goes to zero. In this picture the critical fluctuations, non-magnetic in origin, are associated with the hybridization fluctuation between a broad conduction band and a narrow \(f\)-electron band.

In view of the current interest in KB \cite{3, 8}, it is apt to pose the question if a direct experimental signature of the KB could distinguish it from other possible routes to quantum criticality. The issue is non-trivial partly due to the absence of low-\(T\) angle-resolved photoemission data of the HFs, and partly because standard quantities such as resistivity and specific heat do not measure \(T_K\) directly. For example, the violation of the Wiedemann-Franz (WF) law and the divergence of the Grüneisen ratio (GR) at the QCP have been identified as experimental consequences of KB \cite{5}. However, a true violation of the WF law signifies the absence of electron quasiparticles which need not necessarily be due to \(T_K \rightarrow 0\) physics. An apparent violation of the WF law near a SDW transition \cite{9} can also take place because of inelastic scattering. Similarly, the divergence of the GR is expected for all kinds of QCPs \cite{10}. Consequently, while KB phenomenon implies the violation of the WF law and the divergence of the GR, the converse does not hold. The main purpose of this work is to point out that finite frequency features in the optical conductivity of the HFs, the so-called mid-infrared peak which arise due to inter-band optical transitions, can provide a direct and unique experimental signature of the KB phenomenon.

The mid-infrared peak in the optical conductivity of the HFs has been well-studied experimentally \cite{11} and theoretically \cite{12} in the case where the system is far from any phase instability. The physical origin of the peak is readily understood within a two-band picture of the Kondo effect in a lattice, which describes the formation of the heavy LFL as the hybridization between a conduction \(c\)-band and a narrow \(f\)-band. The inter-band transition takes place between one band with more \(f\)-character and another with more \(c\)-character, and therefore it involves an energy-scale at least of the order of the binding energy \(T_K\) of the composite heavy electrons. Thus the peak position is related to \(T_K\), even though the two quantities may not always be proportional, as we show below.

A crucial aspect of the SDW approach is that the paramagnetic energy scale \(T_K\) remains finite at the QCP. Therefore, for such a transition one does not expect the mid-infrared peak position and the peak height to change significantly while the system is tuned to the QCP by varying the external parameter. In contrast, the KB phenomenon is based upon the possibility of a QCP where \(T_K \rightarrow 0\). Consequently, a hallmark of KB is a significant shift of the inter-band peak position to lower energies, as the system is tuned to the QCP. Furthermore, since at the KB-QCP the \(f\)-band effectively decouples from
the c-band, one expects the inter-band feature to gradually disappear. These qualitative differences can be useful to distinguish experimentally a KB transition from a SDW instability. This motivates us to study how the mid-infrared peak for a heavy LFL varies as the system approaches a KB-QCP.

From the perspective of the charge-dynamics of the f-electrons, the KB-QCP has been identified as an orbital selective Mott transition, where the f-electrons localize and decouple from the metallic environment[13, 14]. Orbital selective localization is a relevant phenomenon in materials with both weakly and strongly correlated bands, including classical Mott insulators (like VO$_2$[15] and V$_2$O$_3$[16]), ruthenates Ca$_{2-x}$Sr$_x$RuO$_4$[17], cobaltates, fullerenes and layered organic superconductors (for a complete list see e.g. Ref. [18]). KB is likely only one of the several possible microscopic mechanisms that leads to orbital selective Mott transition. However it is interesting to conjecture that, besides the HFs, there are other cases where the localization can be described in terms of a vanishing “effective Kondo temperature”. We expect that in such systems the study of the optical conductivity will be equally germane.

Model.— The system we study is described by the Anderson-Heisenberg model which is given by

$$
\mathcal{H} = - \sum_{\langle ij \rangle, \sigma} \left( t_{ij} c_{i \sigma}^\dagger c_{j \sigma} + t_f f_{i \sigma f \sigma} \right) - E_0 \sum_i n_{i \sigma}^f + V_0 \sum_{i, \sigma} \left( c_{i \sigma}^\dagger f_{i \sigma} + \text{h.c.} \right) + U \sum_i n_{i \sigma}^c n_{i \bar{\sigma}}^c + J_H \sum_{\langle ij \rangle} \vec{S}_i \cdot \vec{S}_j.
$$

(1)

Here $t_{ij}$ and $t_f$ ($t_c \gg t_f$) denote hopping of c- and f-electrons respectively, $(i, j)$ are lattice sites (which are, say, nearest and next-nearest neighbours), $E_0$ is the local energy level of the f-band, and $\sigma$ is the spin index. $V_0$ is the hybridization between the bands, $U$ is the on-site Coulomb repulsion in the f-band ($U \gg t_f$), and $J_H \sim (t_f^2)/U$ is a superexchange spin-spin interaction in the f-band. We study the above model in the Kondo limit where the f-electrons form local moments.

The mean field theory is generated by replacing $f_{i \sigma} \rightarrow b_{i \sigma}^\dagger F_{i \sigma}$, where $(b_{i \sigma}^\dagger, b_{i \sigma})$ describe charged holons and $(F_{i \sigma}^\dagger, F_{i \sigma})$ describe spin-1/2 spinons. The large Coulomb repulsion is taken into account by projecting out the double-occupied f-electron states via the constraint $\sum_{\sigma} F_{i \sigma}^\dagger F_{i \sigma} + b_{i \sigma}^\dagger b_{i \bar{\sigma}} = 1$ (the associated Lagrange multiplier renormalizes $E_0$). At the mean field level this model shows a KB quantum phase transition at a critical value $x_c$ of the parameter $x \equiv V_0/J_H$[13]. For $x > x_c$, one obtains an heavy LFL phase where $\langle b_{\bar{\sigma}} \rangle \neq 0$ and the f-electrons participate in band formation, and for $x < x_c$, one obtains a phase with $\langle b_{\bar{\sigma}} \rangle = 0$ where the f-electrons localize and form a non-magnetic uniform spin liquid phase (with $\langle F_{i \sigma}^\dagger F_{j \bar{\sigma}} \rangle \neq 0$).

As the QCP is approached from the LFL side, the lattice Kondo temperature $T_K \approx \pi (b^2 V_0^2/\nu_0) \rightarrow 0$, where $\nu_0$ is the bare density of states of the c-band. We write the spinon bandwidth as $\alpha t_c \sim ((b^2 t_f + J_H)$, and consider $\alpha \ll 1$ as a small parameter of the theory.

We use linear response theory to study the $T = 0$ optical conductivity of the system as it approaches the QCP from the LFL side. We ignore the f-band contribution to the current operator and write it as $\hat{J}_f \approx \sum_{\mathbf{k}} (\nabla_{\mathbf{k}} \epsilon_{\mathbf{k}}) c_{\mathbf{k} \sigma}^\dagger c_{\mathbf{k} \bar{\sigma}}$, where $\epsilon_{\mathbf{k}}$ is the bare dispersion of the c-band, and $\mu$ denotes spatial direction. This does not change our results at a qualitative level, and can be formally justified as an expansion in $t_c/t_f$. Furthermore, we write $|\nabla_{\mathbf{k}} \epsilon_{\mathbf{k}}| \approx v_F$, the Fermi velocity of the c-band, and we ignore the vertex corrections to the current-current correlator. The optical conductivity $\sigma_1(\Omega)$ is the real part of the frequency dependent conductivity tensor, which can be written as (for $\Omega \geq 0$, and setting $\hbar = 1$)

$$
\sigma_1(\Omega) \approx \frac{2 e^2 v_F^2}{\pi \Omega d} \int_{-\Omega}^{0} d\omega \sum_{\mathbf{k}} \text{Im} G_{c R}^R(\omega, \mathbf{k}) \text{Im} G_{c R}^R(\omega + \Omega, \mathbf{k}),
$$

(2)

where $R$ denotes the retarded function. Within the mean field theory the Green’s function for the c-electrons is $G_{c R}^R(\omega, \mathbf{k})^{-1} = \omega - \epsilon_{\mathbf{k}} + i/2\tau_c - \Sigma_{c R}^R(\omega, \mathbf{k})$, $\Sigma_{c R}^R(\omega, \mathbf{k}) = V^2/|\omega - \epsilon_{\mathbf{k}} + i/2\tau_F|$. $V = \langle b \rangle V_0$ is the effective hybridization between the c-band and the F-band of the spinons, $\epsilon_{\mathbf{k}}^0$ is the dispersion of the spinons whose band-width is $\alpha t_c$, and $\tau_c$ and $\tau_F$ mimic the elastic scattering due to the presence of impurities. In principle, $\alpha t_c$ is $V$ dependent, but, for simplicity, we treat it as a constant.

We adopt few simplifications in order to perform the calculations analytically: (i) the c-band dispersion is linearized $\epsilon_{\mathbf{k}} = v_F (k - k_F)$, where $k_F$ is the Fermi wave-vector of the c-band; (ii) the mildly dispersive F-band is replaced by a flat level $\epsilon_{\mathbf{k}}^0 \approx \epsilon_{\mathbf{k}}^F \equiv E_x$ [Fig. 1(a)]. Here $E_x \approx \alpha v_F (k_F - k_{F0})$, where $k_{F0}$ is the Fermi wave-vector of the F-band, and $k_F0 < k_F$. The latter approximation is equivalent to ignoring all non-singular $\alpha$ dependence in Eq. (2). The microscopic non-universal parameter $E_x$, which is the energy necessary for a momentum conserving inter-band transition from the Fermi surface of the c-band to the F-band, becomes an important energy-scale in the limit where the effective hybridization $V$ is small (see below). We do not expect our results to depend on the details of the band-dispersions. To demonstrate this explicitly, we also evaluate numerically $\sigma_1(\Omega)$ in Eq. (2) by taking the continuum limit of the model. We use parabolic dispersions $\epsilon_{\mathbf{k}} = (k^2 - k^2_F)/(2m)$ and $\epsilon_{\mathbf{k}}^0 = (k^2 - k^2_{F0})/(2m_0)$, set the energy scale by $\epsilon_F \equiv k^2_F/(2m) = 1$, and choose $E_x = \epsilon_F/100$, $m/m_0 = \alpha = 0.25$. We show that at a qualitative level the numerical and approximate analytical results match well [Figs. 2, 3], and this validates our physical conclusions.

Results.— Let us for the moment ignore the finite life-
which is finite provided \( \alpha \), conduction and spinon bands (with respective Fermi vectors \( k_F \) and \( k_{F0} \)). \( E_x = \epsilon_k^c \) is a microscopic energy-scale. \( E_{1,2k} \) are the hybridized bands, \( V \) is the effective hybridization and \( \Omega_c = 2V \) is the minimal inter-band transition-energy (corresponds to momenta for which \( \epsilon_k = \epsilon_k^c \)). (b) For \( V > E_x \), the optical conductivity peak is at \( \Omega_c^0 = \Omega_c \). For \( V < E_x \) the \( \Omega_c \)-transition is forbidden at \( T = 0 \), (since \( E_{1k} > 0 \) is above the chemical potential). The peak is at \( \Omega_c^p = V^2/E_x + E_x \).

![FIG. 1: (color online). (a) \( \epsilon_k \) and \( \epsilon_k^c \), are the bare conduction and spinon bands (with respective Fermi vectors \( k_F \) and \( k_{F0} \)). \( E_x = \epsilon_k^c \) is a microscopic energy-scale. \( E_{1,2k} \) are the hybridized bands, \( V \) is the effective hybridization and \( \Omega_c = 2V \) is the minimal inter-band transition-energy (corresponds to momenta for which \( \epsilon_k = \epsilon_k^c \)). (b) For \( V > E_x \), the optical conductivity peak is at \( \Omega_c^0 = \Omega_c \). For \( V < E_x \) the \( \Omega_c \)-transition is forbidden at \( T = 0 \), (since \( E_{1k} > 0 \) is above the chemical potential). The peak is at \( \Omega_c^p = V^2/E_x + E_x \).](image)

In order to include effects of finite lifetime of the \( \sigma_1(\Omega) \) corresponds to momenta for which \( \epsilon_k = \epsilon_k^c \). This corresponds to momenta where the bare bands \( \epsilon_k \) and \( \epsilon_k^c \) cross [Fig. 1(a)]. For this process \( \epsilon_k^c/\epsilon_k \approx 1/4 \) is maximal, and it reduces for inter-band transitions with frequencies larger than \( \Omega_c \).

In order to include effects of finite lifetime of the \( \epsilon \)-electrons (we find that finite \( \tau_F \) is of secondary importance, and we ignore it in the analytic evaluation), it is more convenient to replace the momentum sum in Eq. (2) by an energy integral, which can be performed by the method of contours for the simplified case of a linearized \( c \)-band and a non-dispersive \( F \)-band. This gives

\[
\sigma_1(\Omega) \approx \frac{\Omega_c^2}{\Omega} \text{Im} \int_{-\infty}^{\infty} \frac{d\omega}{\Omega + i/\tau_c + \Sigma_c^A(\omega) - \Sigma_c^R(\omega + \Omega)}.,
\]

where \( \Omega_c^2 = ne^2/m \), \( n \) being the \( c \)-electron density. As \( V \) is reduced we find two different regimes (Figs. 2 and 3).

![FIG. 2: (color online). Evolution of the optical conductivity \( \sigma_1(\Omega) \) with effective hybridization \( V \) ranging from 0.3 \( \times \) 10\(^{-2} \)\( \epsilon_F \) (left) to 9 \( \times \) 10\(^{-2} \)\( \epsilon_F \) (right). \( \sigma_1(\Omega) \) is expressed in unit of \( \epsilon_0 = e^2/(\pi m \epsilon_F) \). The solid lines (Eq. 2) are obtained using parabolic bands (\( \alpha = 0.25 \) and \( 1/\tau_c F \approx 10^{-3} \epsilon_F \)). The dot-dash lines (Eq. 3) are obtained using a linearized \( c \)-band and a non-dispersive \( F \)-band. In both cases, \( V \) is reduced, the peak heights first increase for \( V \gg E_x = \epsilon_F/100 \), and then diminish for \( V \ll E_x \). For clarity the plots are truncated to exclude the Drude feature.](image)
(ii) $V < E_x$. This new regime, which has not been studied earlier, is relevant when the system is close to the KB-QCP. We find that $\Omega = \Omega_c$ corresponds to $E_{1k} = E_x - V > 0$, which implies that an inter-band transition at this frequency is not possible at $T = 0$. The threshold frequency is $\Omega_p^c \equiv V^2 / E_x + E_x > \Omega_c$, and this corresponds to an inter-band transition from the Fermi surface of the lower hybridized band [Fig. 3(b)]. In the vicinity of the threshold we get

$$\sigma_1(\Omega \gtrsim V^2 / E_x + E_x) \approx \frac{\pi \Omega_c^2}{4 \Omega_p^c \sqrt{\Omega^2 - \Omega_c^2}}.$$

As before, neglecting the finite lifetime, the peak-position is at the threshold frequency, and we get [Fig. 3(a)]

$$\Omega_p \approx \Omega_p^c = V^2 / E_x + E_x.$$  

Therefore, $\Omega_p$ is no longer a direct measure of $T_K$, and in particular it stays finite ($\Omega_p \rightarrow E_x$ as $T_K \rightarrow 0$) at the QCP. Also, in this regime $\Omega_p$ varies little with $V$, which may not be discernible whenever $\Omega_p$ cannot be very well-resolved. Since $\Omega_p \neq \Omega_c$, the corresponding matrix element $t_{nk}^2 \Omega^2$ decreases from the maximal value 1/4 as $V$ is reduced. As a result the peak-value $\sigma_1(\Omega_p)$ decreases rapidly [Fig. 2 and Fig. 3(b)], and the peak vanishes at the QCP indicating the gradual decoupling of the $f$-electrons from the conduction band. Note that if $1/\tau_c \gg E_x$, the inter-band feature is eventually masked by the Drude peak.

Next we discuss the limitations of our results. These are based on model calculations and mean field considerations giving more prominent inter-band feature than those observed experimentally. Furthermore, the fluctuations are non-negligible near the QCP, and the crucial question is whether this only smears the feature further or washes it out entirely. The issue requires further investigation that is beyond the scope of the current paper. However, even if the feature is washed out very near the QCP, any observation of significant shift of the peak position towards lower energy will have demonstrated that the physics of these systems is richer than what can be captured by SDW theory.

**Conclusion.**— We studied the finite-frequency inter-band transition peak in the optical conductivity of a heavy fermion system near a Kondo breakdown type of quantum critical point, where the lattice Kondo temperature vanishes. As the system approaches the QCP from the heavy Fermi liquid side, we find a new cross-over regime where the peak position is related to, but is no longer a direct measure of the lattice Kondo scale. In particular, the peak position moves to lower energies, but remains finite at the QCP. On the other hand, the peak value changes non-monotonically as $T_K \rightarrow 0$, and eventually the peak disappears at the QCP indicating the decoupling of the $f$-electrons from the conduction band. These features are fingerprints of a Kondo breakdown type of QCP, and therefore can be used to distinguish it experimentally from a spin density wave type of QCP.
(2007).
[12] P. Coleman, Phys. Rev. Lett. 59, 1026 (1987); L. Degiorgi, F. B. B. Anders, and G. Grüner, Eur. Phys. J. B 19, 167 (2001); J.H. Shim, K. Haule, and G. Kotliar, Science 318, 1615 (2007); H. Weber, and M. Vojta, Phys. Rev. B 77, 125118 (2008).
[13] C. Pépin, Phys. Rev. Lett. 98, 206401 (2007); Phys. Rev. B 77, 245129 (2008).
[14] L. De Leo, M. Civelli, and G. Kotliar, Phys. Rev. B 77, 075107 (2008); Phys. Rev. Lett. 101, 256404 (2008).
[15] J.B. Goodenough, Prog. Solid State Chem. 5, 145 (1971).
[16] S.Y. Ezhov, V.I. Anisimov, D.I. Khomskii, and G.A. Sawatzky, Phys. Rev. Lett. 83, 4136 (1999).
[17] V.I. Anisimov, I.A. Nekrasov, D.E. Kondakov, T.M. Rice, and M. Sigrist, Eur. Phys. J. B 25, 191 (2002).
[18] Imada, M., A. Fujimori, and Y. Tokura, 1998, Rev. Mod. Phys. 70, 1039; see e.g. sec. III.3 in G. Kotliar, S. Y. Savrasov, K. Haule, V. S. Oudovenko, O. Parcollet, and C. A. Marianetti, Rev. Mod. Phys. 78, 865 (2006).