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Optically probing the Kondo resonance in $YbIn_{1-x}Ag_xCu_4$

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Abstract

We report the infrared charge dynamics of $YbIn_{1-x}Ag_xCu_4$ in a series of x- and T-dependent reflectivity measurements. Within the low-temperature coherent phase, characteristics of a low energy (0.25 eV) excitation appear to be controlled by the same Kondo scale, T_K , which is relevant to the magnetic response (Phys. Rev. B 56 (1997) 7993). We show that the observed T_K dependence of the frequency and strength of this excitation is consistent with an interpretation in terms of a coherent-to-incoherent quasiparticle transition between the correlation-induced bands of the periodic Anderson model (Phys. Rev. Lett. 92 (2004) 186405). Additionally, we consider an interpretation of xdependence in the higher frequency optical response in terms of a rigid band structure picture where the filling is depleted by Ag doping, augmented by a Kondo resonance which tracks the x-dependent Fermi level. In addition to the properties of the low-temperature phase, our results also encompass the anomalous temperature dependent behavior in the vicinity of the electronic phase transition at low doping (x < 0.3). © 2005 Elsevier B.V. All rights reserved.

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Investigation of the implications of a Kondo resonance in the excitation spectrum of localmoment magnetic metals continues as current problems surrounding the metallic-magnetic phase boundary inform our thinking about quantum matter. In this report, we focus on a striking midinfrared excitation apparent in the optical conductivity of $YbIn_{1-x}Ag_xCu_4$ and show that aspects of the optical signature of this excitation realize scaling relations with the low-temperature Kondo scale T_K . In addition, the measured trends are in agreement with predictions of the periodic

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Anderson model (PAM). These observations give convincing evidence that the mid-infrared excitation found in the coherent phase of this correlated electron system is a manifestation of the existence of a Kondo resonance near $E_{\rm F}$.

YbInCu₄ features a first-order electronic phase transition from a high-temperature phase, characterized by free-moment magnetism and semimetallic DC transport behavior, into a strongly hybridized metallic phase for $T \leq T_v \simeq 42$ K. Substitution of Ag for In drives upward the valence transition temperature $(\partial T_v/\partial x > 0)$, but also tunes a crossover within the low-temperature phase from the mixed-valent (YbInCu₄, $n_f \simeq 0.8$) toward the regime of moderately heavy fermionic behavior (YbAgCu₄, $n_f \simeq 0.93$).

A microscopic description and understanding of of the valence transition in the origins YbIn_{1-x}Ag_xCu₄ remains elusive, although progress has been made in addressing aspects of the transition, including phenomenology of susceptibility [1], transport [3], the single particle spectrum [4], thermodynamics [5], and aspects of the magnetic field-temperature phase diagram [6]. Recent evidence of a ferromagnetic phase [7] near the $T_v \rightarrow 0$ region of the phase diagram raises new questions concerning the mechanistic description of the valence transition. In addition to our investigations surrounding optical response of the Kondo resonance, we are able to extract several higher energy scales from our optical measurements which could provide direction toward a theoretical description of the valence transition beyond the phenomenological level.

In order to examine the correlated electron behavior of this system, we have made reflectivity measurements of $YbIn_{1-x}Ag_xCu_4$ over a range of temperatures, frequencies, and Ag concentrations. The measured reflectivity spectra were appended with carefully chosen terminations above and below the range of measured data in order to make a complete Kramers–Kronig transform and derive the optical response functions [2,8].

We have previously reported [8] the presence of a mid-infrared (2000 cm⁻¹) feature in the lowtemperature optical conductivity of YbInCu₄. The absence of this feature in the high-temperature phase of YbInCu₄, where $T_{\rm K}$ is negligibly small, leads one to suspect there is a connection with the correlated electron physics of hybridization. Fig. 1 shows an extension of this work with the detailed low-temperature *x*-development of this feature in YbIn_{1-x}Ag_xCu₄. The 2000 cm⁻¹ feature exhibits non-trivial systematic trends which we compare to the *x*-dependent Kondo temperature, $T_{\rm K}$ [1,2].

To explore this possibility systematically, we quantify the strength and frequency of this excitation and compare these spectroscopic quantities to the Kondo temperature $T_{\rm K}$ appropriate to the low-temperature magnetic screening effect [1]. The variation of both $T_{\rm K}$ and the peak position ω_0 of the 2000 cm⁻¹ feature is consistent with a $\omega_0 \propto T_{\rm K}^{0.5}$ scaling. This form arises from theoretical considerations of the periodic Anderson model, when ω_0 is interpreted as the direct gap for quasiparticle excitations (see Fig. 1). In the PAM, this corresponds to an energy $\omega_0 \sim 2\tilde{V}$, where \tilde{V} is the hybridization renormalized by the f-electron on-site repulsion.

Fig. 1 shows that in addition to the peak frequency scaling, there is a correspondence between the Kondo temperature and the strength n of the 2000 cm⁻¹ feature, quantified in this case as the integral of $\sigma_1(\omega)$ between 40 and 4000 cm⁻¹. This correspondence is discussed at length in Ref. [2] where an interpretation in terms of the PAM is given. In this work an idealized line shape is generated by simple considerations in a Fermi's Golden Rule calculation of the transition rate



Fig. 1. Main Panel: The optical conductivity of $YbIn_{1-x}Ag_xCu_4$. Inset: The peak frequency ω_0 and the integral of $\sigma_1(\omega)$ from $\omega = 40-4000 \text{ cm}^{-1}$ plotted against T_K .

from the coherent to incoherent quasiparticle bands. Based on this treatment we conclude that the measured magnitude and direction of the dependence of n on $T_{\rm K}$ is commensurate with PAM predictions.

In a simplified impurity model approach, one can estimate the bare hybridization in this context by noting that the measured mass $m^* = 1/\sqrt{z}$ and dressed hybridization are related by $\tilde{V}^2 = zV^2$. For YbIn_{1-x}Ag_xCu₄, the dressed hybridization is in the range $\tilde{V} \simeq 50-100$ meV, with associated effective masses in the range $m^* \sim 50-10 m_B$. The bare hybridization V deduced from these considerations is of the order of 0.28 eV.

Systematic changes are also observed in the 1.2 eV feature as x is increased and Ag is substituted for In, removing a total of 2 electrons/Yb and thereby lowering the Fermi level. In concert, the low energy edge of the 1.2 eV feature moves approximately 0.5 eV. In a rigid band interpretation of this shift, one obtains $\langle \text{DOS}(E_{\rm F}) \rangle \simeq 2e^{-}/(0.5 \text{ eV}) = 4e^{-}/eV$ as the mean density of states in this region of the band structure. This is a typical value for *d*-derived (Ag, Cu, In) electronic bands.

These parameters can be combined to deduce an impurity model f-level broadening $\Delta = \pi V^2 \text{DOS}(E_F) \sim 1.25 \text{ eV}$, suggesting that the hybridization effects are sizable enough to control the physics in this system.

We have thus made a quantitative connection between existing correlated electron theory through comparisons of the two-particle spectrum of low-temperature YbIn_{1-x}Ag_xCu₄ with local moment models, thereby suggesting parameter values of the effective model Hamiltonian for the low temperature phase as well as identifying the phenomenology associated with a Kondo resonance excitation in this compound.

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