RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 62, NUMBER 8 15 AUGUST 2000-II

# Optical study of the electronic phase transition of strongly correlated $YbInCu_4$

Sean R. Garner, Jason N. Hancock, Yvonne W. Rodriguez, and Zack Schlesinger
*Department of Physics, University of California, Santa Cruz, California 95064*

Benno Bucher
*Department of Physics, ITR, Oberseestrasse 10, 8640 Rapperswil, Switzerland*

Zach Fisk
*Department of Physics and NHMFL, Florida State University, Talahassee, Florida 32310*

John L. Sarrao
*Los Alamos National Laboratory, Los Alamos, New Mexico 84545*

(Received 17 February 2000)

Infrared, visible, and near-UV reflectivity measurements are used to obtain conductivity as a function of temperature and frequency in $YbInCu_4$, which exhibits an isostructural phase-transition into a mixed-valent phase below $T_v \simeq 42$ K. In addition to a gradual loss of spectral weight with decreasing temperature extending up to 1.5 eV, a sharp resonance appears at 0.25 eV in the mixed-valent phase. This feature can be described in terms of excitations into the Kondo (Abrikosov-Suhl) resonance, and, like the sudden reduction of resistivity, provides a direct reflection of the onset of coherence in this strongly correlated electron system.

Phenomena in the field of strongly correlated electron systems can often be described in terms of either a Mott-Hubbard model or a periodic Anderson model[1] (PAM). $YbInCu_4$ is difficult to classify in this manner. Its low temperature properties appear to be consistent with a PAM approach—it is mixed-valent with an enhanced carrier mass and a correspondingly enhanced electronic specific heat and Pauli spin susceptibility. However, it exhibits a phase transition at 42 K to a high-temperature state with integer valence and conventional electronic mass. This transition is beyond the scope of the periodic Anderson model, which generally exhibits gradual changes (crossovers) in its temperature dependence.[1] It is, however, reminiscent of Mott-Hubbard physics, in which a phase transition associated with a strong Coulomb repulsion term in the Hamiltonian can occur. In this paper we present spectroscopic studies of the phase transition and the low-temperature state of $YbInCu_4$ which show the emergence below the transition of a well-defined mode at an energy of about 0.25 eV. Possible interpretations of this electronic excitation, and the question of what Hamiltonian is required to capture the essential physics of $YbInCu_4$ are discussed.

The presence of local magnetic moments in metallic systems is associated with a variety of interesting phenomena, including the Kondo effect, heavy-fermion physics, and mixed-valence.[1] In certain cases, an isostructural first-order transition occurs at which a discontinuous change in valence and volume is accompanied by an abrupt disappearance of the local moment.[2–6] The transition to such a state provides an opportunity to explore fundamental issues associated with magnetic moments in metals, moment compensation, Kondo singlet formation, and mixed-valence phenomena.

At the $\gamma$-$\alpha$ transition[2] of Ce a valence change from about 3 to 3.2 occurs in concert with a volume reduction of about 15% at $T_v \sim 200$ K. This transition has been discussed in terms of a Kondo-volume-collapse model.[3] In that model, a reduced lattice constant in the low-temperature phase is associated with an increase in the hybridization interaction between local moment and conduction electron states, which leads to moment disappearance due to the formation of a Kondo-singlet state in which the *f*-level moments are compensated due to a Kondo-like screening by conduction electrons.[1]

$YbInCu_4$ exhibits an isostructural transition to a mixed-valent ground state at which the volume change is almost negligible ($\simeq 0.5\%$).[7–11] At this transition the Yb valence decreases from $\sim 3$ to $\sim 2.85$, and the local moment, present in the high-temperature state, vanishes abruptly. The Kondo scale, $T_K$, which is a measure of the strength of the hybridization interaction, increase abruptly from $T_K \approx 25$ K above the transition to $T_K \approx 400$ K below the transition for reasons which are not at all clear.

In this paper, we focus on changes in the infrared conductivity of $YbInCu_4$ associated with the transition to the mixed-valent state. The abrupt increase of the Kondo scale below $T_v$ can allow us to identify key features of the Kondo state, and thus shed light on fundamental phenomena of Anderson lattice systems. This work is complementary to previous optical work which addressed the relationship between spectral features and band-structure calculations.[12–15] It is also possible that the very high purity of present samples[8,16] allows features to be observed that could not be readily discerned in earlier work.

The samples used in these experiments are high-quality single crystals grown from an In-Cu flux.[9] For these samples a sharp transition occurs at about 42 K in the absence of strain. At the transition the volume increases by about 0.5% as the sample is cooled, and the susceptibility and resistivity drop abruptly by an order of magnitude. Thermal cycling

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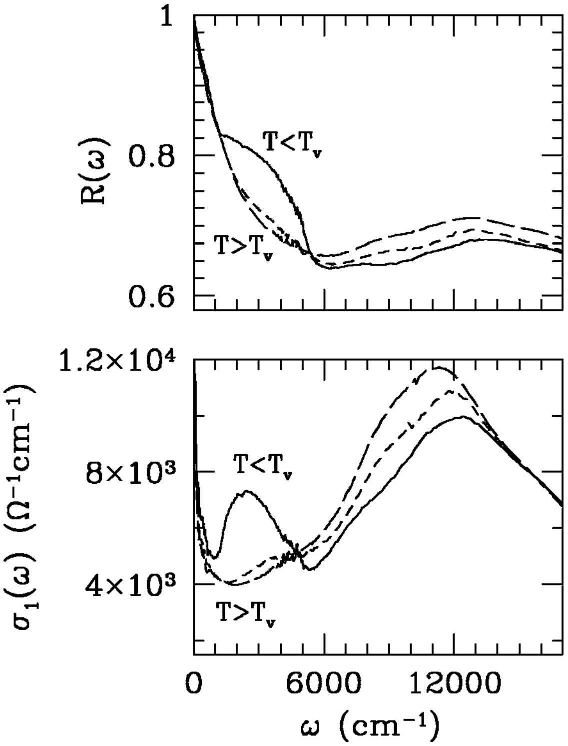

FIG. 1. The reflectivity and the real part of the conductivity at low frequency are shown for $YbInCu_4$ at $T = 250$ K (long dashes), 55 K (shorter dashes), and 20 K (solid line). Gradual reduction of spectral weight with cooling occurs in the vicinity of 10 000 $cm^{-1}$ (1 eV). A well-defined resonance appears at 2000 $cm^{-1}$ (1/4 eV) in the low temperature Kondo state.

tends to induce strain in the samples, which can broaden the transition and move it to higher temperature.[9] Infrared and optical measurements are performed using a combination of Fourier transform and grating spectrometers to cover the range from 50 to 50 000 $cm^{-1}$. In these measurements we have gone to great efforts to measure in all ranges before going through the transition to avoid disorder effects influencing the infrared data significantly. The conductivity as a function of frequency is obtained from a Kramers-Kronig transform of the reflectivity data. For the purpose of performing this transform, the measured reflectivity is extended from 50 000 to 200 000 $cm^{-1}$ as a constant, and above that it is made to decrease like $1/\omega^2$. At low frequency a Hagen-Rubens termination is attached to the data. In the region of the actual data, the conductivity is insensitive to the details of these terminations.

Figure 1 shows reflectivity and conductivity up to 15 000 $cm^{-1}$. These data show the emergence of a prominent resonance in the mixed-valent (low-$T$) state near 2000 $cm^{-1}$ ($\simeq 1/4$ eV). The spectral sharpness and the abrupt appearance of this feature as a function of temperature are striking.

In addition, the data indicate the persistence of significant temperature dependence to relatively high frequency (compared to $T$ or $T_K$) in $YbInCu_4$. In the frequency range from about 5000 to 12 000 $cm^{-1}$, $\sigma_1(\omega)$ decreases substantially as $T$ is reduced both above and below $T_v$. Recent theoretical[17,18] and experimental work[19–23] has explored the phenomenology and possible origins of such high energy spectral weight shifts in strongly correlated systems (involving energies vastly larger than $k_BT$ and $k_BT_K$).

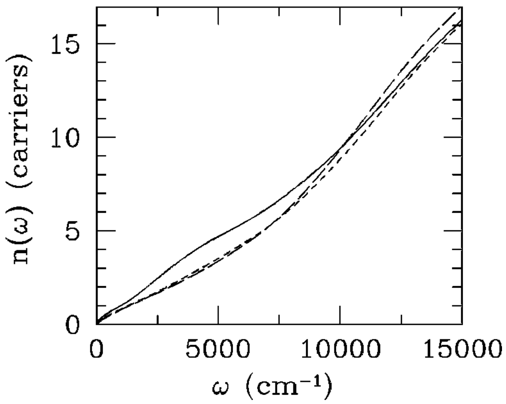

FIG. 2. Spectral weight, the integral of $\sigma_1(\omega)$ as a function of frequency from 0 to $\omega$, is shown as a function of $\omega$ at $T = 250$ K (long dashes), 55 K (shorter dashes), and 20 K (solid line). The units for the vertical axis, expressed in terms of carriers per Yb atom, are established by the assumption of a band mass of 4.

Figure 2 shows spectral weight, which is the indefinite integral of $\sigma_1(\omega)$,

$$n(\omega) = \frac{m}{\pi e^2}\int_0^{\omega} \sigma_1(\omega')\,d\omega',$$

as a function of frequency. In this figure (and in Fig. 1) we see that there is a net loss of spectral weight as the temperature is lowered from 250 to 55 K. The loss amounts to about 10% of the strength of the broad mode centered around 12 000 $cm^{-1}$, and corresponds to $\sim 1.5$ carriers/Yb atom with the reasonable assumption of a band mass of four times the free-electron mass. Since spectral weight is ultimately conserved (if one integrates to high enough frequency[24]), these data imply that it must be displaced to still higher frequency (above 16 000 $cm^{-1} \simeq 2$ eV) as $T$ is reduced from 250 to 55 K.

On the other hand, the coalescence of the 20 and 55 K curves at the high-frequency end of Fig. 2 indicates that spectral weight is conserved within the frequency range from 0 to 15 000 $cm^{-1}$ as the sample goes through the transition. In particular, the increase in spectral weight associated with the appearance below $T_v$ of the resonance at $\sim 2000$ $cm^{-1}$, which corresponds to about 1.5 carriers/Yb, is balanced by a general reduction of $\sigma_1(\omega)$ up to $\sim 15\,000$ $cm^{-1}$.

Both the starting Hamiltonian and the mechanism that drives the transition to the mixed-valent state remain areas of active research for $YbInCu_4$. With regard to the mechanism, it has been argued that the lattice expansion is clearly too small to directly account for the large change in Kondo temperature at the transition[25] (from $T_K \simeq 25$ to 400 K). The Falikov-Kimball model is capable of producing a quasi-Hubbard-like first-order transition, and may be relevant to high-temperature properties of $YbInCu_4$, however it ignores hybridization, which is certainly important in the low-$T$ state.[26] In the mixed-valent state, where the Kondo scale is large, the dynamics of the periodic Anderson model (PAM) are expected to be relevant.

Within the PAM context, the 1/4 eV excitation can be associated with a quasiparticle interband transition involving

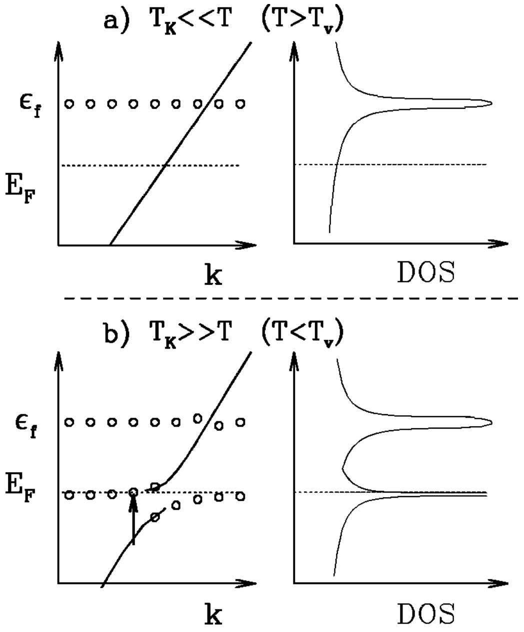

FIG. 3. A generic picture of band dispersion and density-of-states (DOS) for a Kondo-lattice or PAM system (Ref. 1) is shown for the case of a single *f* hole. For the *E* vs *k* plots, solid lines refer to conduction-band states, and circles indicate states with significant *f* character. The dotted line is the Fermi level (chemical potential). (a) For $T_K \ll T$ the *f* level ($\epsilon_f$), associated with the Yb local moments, lies well above the Fermi level ($E_f$) and only a rapidly dispersing conduction band crosses $E_f$. (b) For $T_K \gg T$ a many-body Kondo hybridization creates slowly dispersing states with *f*-like character near $E_f$ (the Abrikosov-Suhl resonance) and an optical transition into such states becomes allowed, as shown by the arrow. We propose that the resonance at 0.25 eV in Fig. 1 may correspond to this transition.

Kondo resonance states near $E_f$,[27,28] as illustrated in Fig. 3, which exist only for $T_K \gg T$. The abrupt change of $T_K$ at the transition and the abrupt appearance of the resonance are consistent with this interpretation. Note that just above the transition $T_K/T \simeq 1/2$, whereas just below the transition $T_K/T \simeq 10$. The study of the phase transition of $\mathrm{YbInCu_4}$, at which $T_K$ changes abruptly by an order of magnitude, can thus allow the first clear identification of this fundamental excitation.

According to model calculations,[27] the energy scale for this excitation involving dynamically generated quasiparticle states at $E_f$ (the Kondo, or Abrikosov-Suhl, resonance) is expected to be $\sim \sqrt{T_K B}$. Here *B* is the conduction bandwidth, which appears because of its connection to how rapidly the conduction band disperses across $E_f$ (cf. Fig. 3). $T_K$ is a measure of the strength of the dynamical hybridization that splits the bands at $E_f$.

Since $T_K \simeq \tilde{V}^2/B$, the infrared determination of this excitation energy provides a measure of the renormalized hybridization, $\tilde{V}$. Using the value $\tilde{V} \simeq 1/4$ eV from our infrared data, along with $T_K \simeq 400$ K ($\simeq 35$ meV), implies a bandwidth of $B \simeq 1.8$ eV, which is reasonable. One can estimate the hybridization broadening, $\Gamma$, using its relationship to $\tilde{V}$, to be $\Gamma \simeq 0.25$ eV. Further, one can use $\Gamma$ in NCA formulas[29] involving $n_f(T)$ along with $L_{III}$ edge measurements of valence[30] to infer that the *f*-level ($\epsilon_f$) is about 0.5 eV away from the chemical potential. These values are all quite reasonable for this mixed-valent system.

Figures 1 and 2 show that the growth of the 0.25 eV mode is associated with a reduction of spectral weight at frequencies up to about 12 000 $\mathrm{cm}^{-1}$ (1.5 eV). This observation—that the growth of the resonance at $\simeq 1/4$ eV comes from a redistribution of spectral weight from essentially the entire bandwidth—may have implications for questions related to exhaustion and the time scales relevant to screening in Kondo lattice systems.[28,31] Does it suggest that conduction electrons further than $k_B T_K$ from the chemical potential are significantly involved in screening in the Kondo lattice? Further work can be expected to address such questions. It is also intriguing to note that an excitation of similar frequency is present in $\mathrm{YbB_{12}}$,[32] for which $T_K \simeq 300$ K, and that related features may also be present in spectra from mixed-valent Ce compounds.[21]

In conclusion, we have examined the infrared conductivity of $\mathrm{YbInCu_4}$, which exhibits an electronic phase-transition (from a magnetic state to a nonmagnetic, mixed-valent state) at $T_v \simeq 42$ K. The most striking feature of the data is the emergence of an electronic excitation at $\sim 0.25$ eV in the low-temperature state, which we discuss in terms of excitations into the dynamically generated Kondo (Abrikosov-Suhl) resonance at the Fermi surface. Like the sudden reduction of resistivity and loss of local moment susceptibility, the emergence of this feature appears to provide a direct signal of the onset of coherence in this strongly correlated electron system.

The authors acknowledge valuable conversations with D. L. Cox, J. W. Allen, J. K. Freericks, S. A. Kivelson, D. H. Lee, and A. P. Young, and technical assistance from T. Lorey, S. L. Hoobler, and P. J. Kostic. Work at UCSC was supported by the NSF through Grant Nos. DMR-97-05442 and DMR-00-71949. Work at Los Alamos was performed under the auspices of the U.S. Department of Energy. NHMFL is supported by the NSF and the state of Florida. Z.F. and J.L.S. also acknowledge partial support from the NSF under Grant No. DMR-9501529. Y.W.R. acknowledges support from the Marilyn C. Davis Endowment for Re-entry Women in Science.

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