Two Energy Scales and Slow Crossover in $YbAl_3$

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Experimental results for the susceptibility, magnetization, specific heat, $4f$ occupation number, Hall effect, and magnetoresistance for single crystals of the intermediate valence (IV) compound $YbAl_3$ show that, in addition to the Kondo temperature scale $T_K \sim 670$ K, there is a low temperature scale $T_{coh} \sim 30-40$ K for the onset of Fermi liquid coherence. Furthermore, the crossover from the low temperature Fermi liquid regime to the high temperature local moment regime is slower than predicted by the Anderson impurity model. We suggest that these effects are generic for IV compounds and we discuss them in terms of the theory of the Anderson lattice.

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The low temperature transport behavior of periodic intermediate valent (IV) and heavy fermion (HF) compounds [1] is fundamentally different from that expected for the Anderson impurity model (AIM), in that it manifests vanishing resistivity (Bloch's law) and an optical conductivity [2] appropriate for renormalized band behavior. Similarly, the $4f$ electrons have a coherent effect on the Fermi surface, as seen in de Haas van Alphen (dHvA) measurements [3]. However, the temperature dependence of the susceptibility, specific heat and $4f$ occupation number, and the energy dependence of the dynamic susceptibility show behavior that is qualitatively very similar to the predictions of the AIM [4,5]. Essentially this is because these properties are dominated by spin/valence fluctuations that are highly local and which exhibit a Lorentzian power spectrum [6] consistent with the AIM.

In this paper we report data for the IV compound $YbAl_3$ and show that these latter properties can differ in at least two ways from the predictions of the AIM. First, the crossover from low temperature Fermi liquid behavior to high temperature local moment behavior is slower (i.e., more gradual) than predicted for the AIM. Second, anomalies (relative to the AIM) in the magnetization, susceptibility, specific heat, and magnetotransport occur below 30–40 K, which is the temperature scale T_{coh} for the onset of coherent Fermi liquid T^2 behavior in the resistivity. We then argue that the existence of a slow crossover and a low temperature scale that is an order of magnitude smaller than the AIM Kondo temperature ($T_{coh} \ll T_K$) may be generic features of IV compounds and we discuss them in terms of the theory of the Anderson lattice.

The samples were single crystals of $YbAl_3$ and $LuAl_3$ grown by the "self-flux" method in excess Al. The susceptibility was measured using a SQUID magnetometer and the specific heat was measured via a relaxation technique. The Hall coefficient was measured in a magnetic field of 1 T using an ac resistance bridge. The high field magnetization and the magnetoresistance were measured at the Los Alamos Pulsed Field Facility of the National High Magnetic Field Laboratory. The $4f$ occupation number $n_f(T)$ was determined from the Yb L_3 x-ray absorption near-edge structure, measured at the Stanford Synchrotron Radiation Laboratory (SSRL); the details of the experiment and the analysis were similar to those discussed earlier [7]. In particular, the Lu L_3 near-edge structure measured for $LuAl_3$ was used as a standard.

In Fig. 1 we plot the susceptibility $\chi(T)$ and the linear coefficient of the $4f$ specific heat, where $\gamma_m = C_m/T$ and $C_m = C(YbAl_3) - C(LuAl_3)$. (For $LuAl_3$ at low temperatures, $C = \gamma T + \beta T^3$ with $\gamma = 4$ mJ/mol K^2 and $\beta = 1.15 \times 10^{-4}$ J/mol K^4, which implies a Debye temperature $\Theta_D = 257$ K.) The broad peaks near 100 K are typical of Yb IV compounds with T_K greater than 500 K (see Fig. 4). However, the low temperature specific heat coefficient displays an upturn below 30 K which saturates at $T = 0$. In addition, the susceptibility increases below 40 K to a peak at 15 K. This was reported earlier by Hiess *et al.* [8] who raised the possibility that it might represent an extrinsic effect due to antiferromagnetic short range ordering of Yb^{3+} impurities. Our argument against this is that the quality of our samples, as measured by the residual resistivity ($0.5 \mu\Omega$ cm), is sufficiently high that dHvA signals are well resolved [9]; the presence of Yb^{3+} impurities should lead to a large residual resistivity. Furthermore, we find that as the residual resistivity increases, the $T = 0$ specific heat maximum decreases and a Curie tail grows

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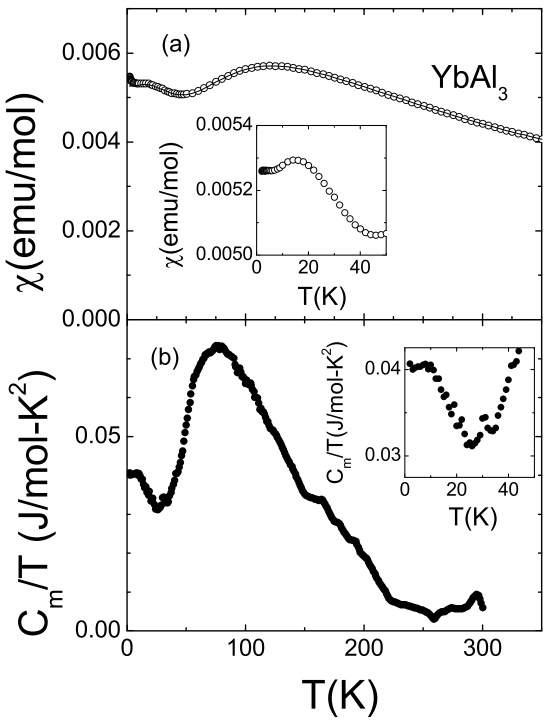

FIG. 1. (a) The magnetic susceptibility $\chi(T)$ and (b) the magnetic contribution to the specific heat coefficient C_m/T for $YbAl_3$. The insets exhibit the low temperature behavior; a small Curie tail has been subtracted from the data in the inset for $\chi(T)$.

in the susceptibility. Hence we associate the low temperature anomalies in susceptibility and specific heat with the purest samples, and assert that they are intrinsic. These anomalies are the basic evidence for the existence of a low temperature scale, $T_{coh} \sim 30-40$ K. The inset of Fig. 3 shows that a T^2 behavior of the resistivity sets in below 30 K, which makes it clear that T_{coh} is the temperature scale for the onset of coherent Fermi liquid behavior.

In Fig. 2a we plot the magnetization versus applied magnetic field at low and high temperature; the solid lines are linear fits to the data. The difference between the linear fits and the data is shown in the inset. At 250 K the magnetization is linear in field up to 60 T; at 4 K the data is linear up to 40 T, above which there is a clear change in slope. We plot the slope $\chi(H)$ for low and high field versus temperature in Fig. 2b. The low field results compare well with the susceptibility measured for $B = 1$ T in the SQUID magnetometer, showing a maximum near 125 K and a second maximum near $T = 0$. In the high field data, the low temperature maximum is absent and the data exhibit the qualitative features expected for an Anderson impurity (see, e.g., Fig. 4b). Clearly, the low temperature susceptibility anomaly is suppressed by magnetic fields greater than $B^* \sim 40$ T. Since $\mu_B B^*$ is of the same order as $k_B T_{coh}$, this effect gives strong confirming evidence for the existence of a new low *energy* scale $k_B T_{coh} \sim 3-4$ meV which is an order of magnitude smaller than the AIM Kondo scale.

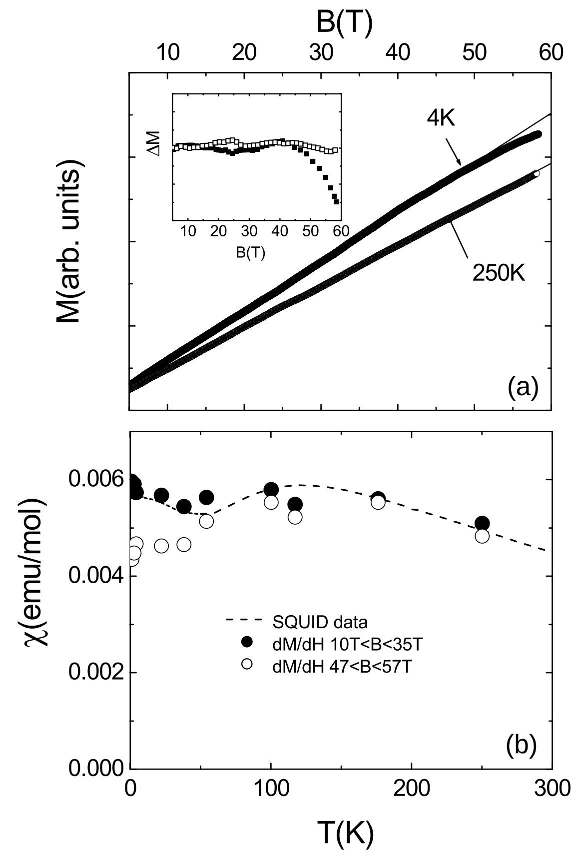

FIG. 2. (a) The magnetization $M(H)$ versus the magnetic field at 4 and 250 K; the solid lines are linear fits to the data in the field range $10 < B < 35$ T. Inset: The difference ΔM between the data and the linear fits. (b) The susceptibility $\chi(T)$ measured at both low ($10 < B < 35$ T) and high ($47 < B < 57$ T) magnetic field. The dashed line is data taken at $B = 1$ T.

Anomalies can also be seen in the magnetotransport (Fig. 3). The Hall coefficient of $LuAl_3$ is temperature independent, as is typical of a metal. The high temperature Hall coefficient of $YbAl_3$ varies with temperature in a manner suggestive of scattering from Yb moments (although the data cannot be fit well with the standard skew scattering formula [10]). Near 50 K the derivative dR_H/dT of the Hall coefficient changes sign. The magnetoresistance (Fig. 3b, inset) follows a B^2 law above 50 K and the magnitude is approximately the same for field parallel and transverse to the current. Below 50 K, the magnetoresistance becomes more nearly linear and the transverse magnetoresistance becomes substantially larger ($\Delta R/R \sim 0.75$) than the parallel magnetoresistance ($\Delta R/R \sim 0.35$) at 2 K. In Fig. 3b we plot $\Delta R/R$ versus Br_0 where $r_0 = R(150K)/R(T)$; this tests Kohler's rule, i.e., that at any temperature $\Delta R/R = Af(Br_0)$ depends only on the product Br_0. The data violate this rule essentially because A varies with T, increasing by a factor of almost 1.5 between 40 and 80 K; this crossover is seen most clearly in the data measured as a function of temperature at a fixed field 17.5 T. These magnetotransport anomalies suggest

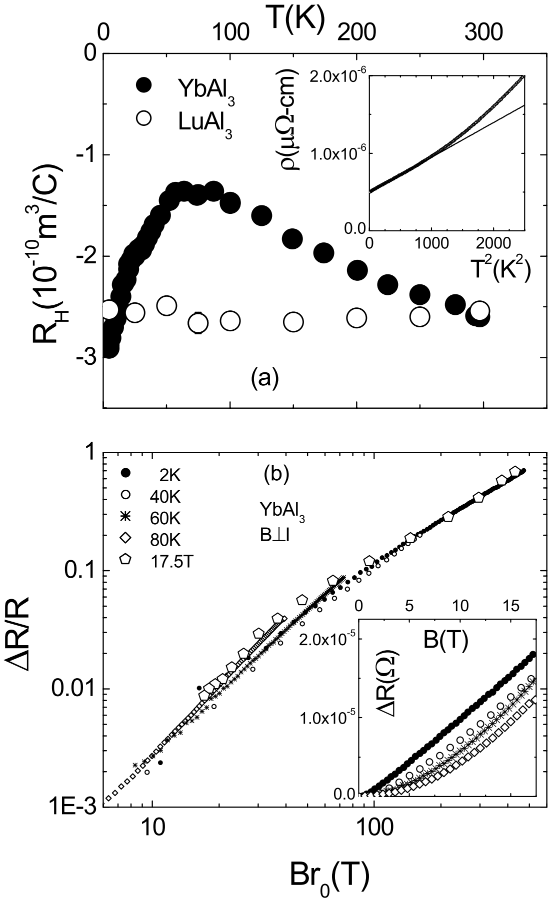

FIG. 3. (a) The Hall coefficient of $YbAl_3$ (closed circles) and $LuAl_3$ (open circles) versus temperature. Inset: the resistivity plotted versus the square of the temperature. (b) Inset: The transverse magnetoresistance $\Delta R = R(H,T) - R(0,T)$ versus magnetic field for four temperatures. Main panel: The relative magnetoresistance $\Delta R/R$ versus Br_0 where $r_0 = R(150\ \mathrm{K}/R(T))$.

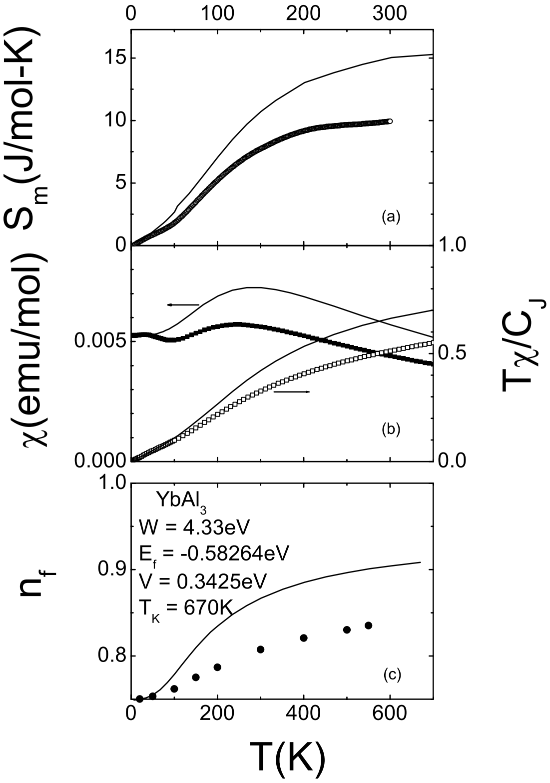

FIG. 4. (a) The $4f$ entropy S_m, (b) the susceptibility $\chi(T)$ (solid symbols), and the effective moment $T\chi/C_J$ (open symbols) where C_J is the $J = 7/2$ Curie constant and (c) the $4f$ occupation number $n_f(T)$ for $YbAl_3$. The symbols are the experimental data and the solid lines are the predictions of the Anderson impurity model (AIM) calculated in the NCA with input parameters as given in the figure.

that the anomalies in χ and C/T may be associated with an alteration of the Fermi surface.

To demonstrate that the crossover from Fermi liquid behavior to local moment behavior is slower than predicted by the AIM we proceed as in our recent paper on $YbXCu_4$ [5]. The calculations were performed using the noncrossing approximation (NCA); the values of the AIM parameters are given in Fig. 4. It is clear from Fig. 4 that the susceptibility, $4f$ occupation number, and $4f$ entropy $S_m = \int dT\, C_m/T$ all *qualitatively* follow the predictions of the AIM. The calculated coefficient of specific heat ($\gamma = 47.8$ mJ/mol K^2) is within 20% of the measured value ($\gamma_m = 40.65$ mJ/mol K^2). Indeed the data even are in accord with the prediction that the entropy $S_m(T)$ approaches the high temperature limit faster than the effective moment $T\chi/C_J$ which in turn evolves more rapidly than $n_f(T)$ (see Fig. 4). Nevertheless, it is also clear that the experimental data for these quantities approach the high temperature limit considerably more slowly than predicted by the AIM theory.

Thus we have demonstrated the existence of a new low energy scale for $YbAl_3$ and we have shown that the crossover to local moment behavior is slower than expected based on the Anderson impurity model. We believe that these effects are generic to IV compounds. We have recently shown [5] that a slow crossover exists in a number of $YbXCu_4$ IV compounds. A number of years ago we gave evidence [11] for a small coherence scale in the IV compound $CePd_3$ based on a low temperature peak in the susceptibility and the extreme sensitivity of the transport behavior to Kondo hole impurities below 50 K. A low temperature anomaly in the Hall coefficient of several Ce compounds also has been observed [10]. Optical conductivity measurements [2,12,13] showed that the temperature scale for these effects is the same as for the renormalization of the effective mass and the onset of the hybridization gap. Hence the anomalies appear to be associated with the onset of the fully renormalized coherent Fermi liquid ground state. Measurement of the infrared optical conductivity is a key future experiment for $YbAl_3$.

An open issue is the condition under which these anomalies occur. One possibility is low background conduction electron density. In $CePd_3$ the carrier density is less than 0.1/atom [2]. For $YbXCu_4$ we found [5] that the slow crossover did not correlate either with the degree of mixed valence or with the Kondo temperature, but it did correlate with a low background conduction electron density as calculated from the Hall coefficient of LuXCu$_4$ in a one-band approximation. Using a similar approximation we deduce from the Hall coefficient of LuAl$_3$ (Fig. 3a) that $n_c \sim 0.5/\text{atom}$. This is as low as in those YbXCu$_4$ compounds where strong deviations from the AIM are observed.

Indeed, recent theoretical work [14,15] on the Anderson lattice predicts both a slow crossover and a low temperature coherence scale in the limit of low conduction electron density. However, the theoretical work to date has been performed only in the Kondo limit. We have examined the extension of the slave boson mean field theory for the Anderson lattice to the case $n_f < 1$ and $n_c < 1$ relevant to YbAl$_3$, where n_f is the number of holes in the $(2J + 1)$ fold degenerate f level. Following Millis and Lee [16], we define the Kondo temperature $k_B T_K$ as the energy of the renormalized f level relative to the Fermi energy. The coherence scale is defined as the renormalized (quasiparticle) $T = 0$ bandwidth which for a background conduction band density of states ρ is given by $k_B T_{\text{coh}} = \rho \tilde{V}^2$ where $\tilde{V}$ is the renormalized hybridization $\tilde{V} = \sqrt{1 - n_f}\, V$. In the limit $(\rho \tilde{V})^2 \ll n_c n_f/(2J + 1)^2$ we find that $T_{\text{coh}}/T_K = n_f/(2J + 1)$ independent of n_c. For YbAl$_3$ this means that T_{coh} should be an order of magnitude smaller than T_K, in qualitative agreement with the data.

In any case we assert that the two energy scales and slow crossover predicted by recent theory are features of our data, that they are generic for IV compounds, and that these effects show some correlation with a standard measure of the conduction electron density.

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