Two energy scales and slow crossover in $YbAl_3$

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$YbAl_3$ is an intermediate valence (IV) compound with a large Kondo temperature scale T_K ~ 670K for the crossover from local moment behavior to nonmagnetic behavior. Above 30-50K, the behavior is that of uncorrelated Kondo impurities and can be understood qualitatively in terms of the Anderson impurity model (AIM). Below a coherence temperature T_{coh} ~ 30-50K the d.c. and optical conductivity indicate that the system enters a Fermi liquid (FL) ground state in which the effective masses (as determined by dHvA) are large (m^* ~ 15-25m_e). Anomalies in the susceptibility, specific heat, magnetotransport and spin fluctuation spectra occur for $T < T_{coh}$. The dHvA masses and the susceptibility anomaly are suppressed by application of a magnetic field $H > 40T \sim k_B T_{coh}/\mu_B$. In addition, the crossover from the nonmagnetic to the local moment regime is slower than predicted by the AIM. We discuss these results in terms of the Anderson lattice model, with consideration given to the role of low electron density.

Collaborators

- Los Alamos: Joe Thompson $\chi(T)$
  John Sarrao Crystal growth
  Mike Hundley R_H, $\rho(H)$
- NHMFL/LANL Alex Lacerda $\rho(H)$
- LBL ` Corwin Booth $n_f(T)$ (L_3 XRA)
- Shizuoka U. Takao Ebihara Crystal growth; dHvA
- U. Nevada, Las Vegas Andrew Cornelius M(H); dHvA
- Temple U. Peter Riseborough Theory (NCA)
- IPNS @ ANL Ray Osborn $\chi''(\omega)$ (neutron scattering)
- HFBR @ BNL Steve Shapiro ••• ••• •••

Intermediate Valence (IV) Compounds

Intermediate valence compounds: $CePd_3$, α-Ce, $YbAgCu_4$, $YbAl_3$, etc.

Archetypal class of systems subject to electron-electron correlations.

Basic Physics: Highly localized 4f electron
degenerate with and hybridizing with conduction electrons
with strong on-site Coulomb interactions between 4f electrons

More complex than TM's (e.g. Pd) or 3D one-band Hubbard Model

Less complex than TM oxides (e.g. high-T_c) which have:
Multiple bands, 2D character, possible hidden order,
quantum critical point (QCP)

Comparison to Heavy Fermions (HF)

HF	***IV***
CF doublet ground state:	CF unimportant:
$T_K < T_{cf}$	$T_K > T_{cf}$
$N_J = 2J + 1 = 2$	$N_J = 6$ (Ce); 8 (Yb)
Proximity to QCP:	Proximity to Kondo Insulator:
AF correlations	Hybridization gap
	No AF correlations
Low symmetry (tet, hex)	High symmetry (cubic)
Anisotropy (sometimes 2D effects)	Isotropic (3D)

Basic low temperature properties of IV compounds

Intermediate Valence (IV) = Nonintegral valence

Partial occupation of the 4f shell

Yb:	$(5d6s)^3\ 4f^{13}$	$n_f = 1$	trivalent
	$(5d6s)^2\ 4f^{14}$	$n_f = 0$	divalent
$YbAl_3$	$(5d6s)^{2.8}\ 4f^{13.2}$	$n_f = 0.8$	IV

Fermi Liquid with enhanced effective mass:

T-linear specific heat for Fermi liquid:

$$\gamma = \{\pi^2 k_B^2 N_A Z/(3 h^3 \pi^2 N/V)^{2/3})\}\ m^*$$

For simple metals (e.g. K): $\gamma = 2\text{mJ/mol-K}^2$ so $m^* = 1.25m_e$

For IV $YbAl_3$: $\gamma = 40\text{mJ/mol-K}^2$ so $m^* \sim 20m_e$

$\Rightarrow$ "(Moderately) **Heavy Fermion**"

Pauli paramagnetism

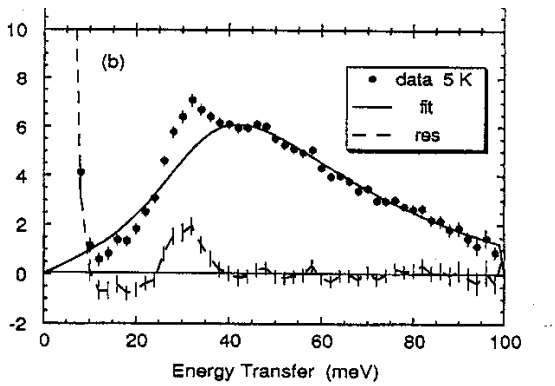
$YbAl_3$: $\chi(0) = 0.04\text{emu/mol}$

Spin Fluctuation Spectra
(Neutron Scattering)

It is known from studies of single crystals of $YbInCu_4$ (Lawrence, Shapiro et al, PRB55 (1997) 14467) that the spin fluctuations in IV compounds show very little Q- dependence and the magnetic scattering exhibits a Lorentzian power spectrum.

$$\chi''(Q,E) = (n(E)+1)\, f^2(Q)\, \chi'(Q)\, E\; P(E)$$
$$P(E) = (\Gamma/2)\, \{(E - E_0)^2 + \Gamma^2)^{-1} + (E + E_0)^2 + \Gamma^2)^{-1}\}$$
$$f^2(Q): \text{ 4f form factor}$$

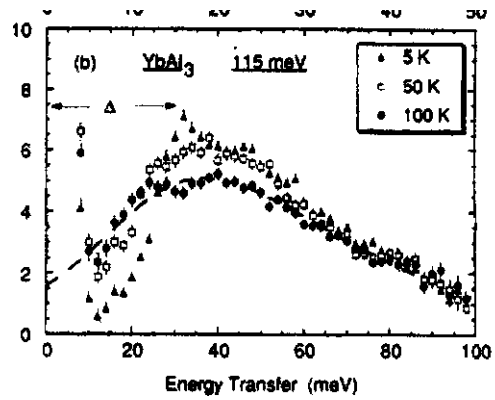
Q-independent, broad Lorentzian response $\Rightarrow$
Primary excitation is a local, highly damped spin fluctuation (oscillation) at characteristic energy $E_0 = k_B T_0$



Murani, PRB 50 (1994) 9882

For $YbAl_3$ the parameters of the low temperature Lorentzian are:
$E_0 = 40$meV and $\Gamma = 25$meV
(Solid line)

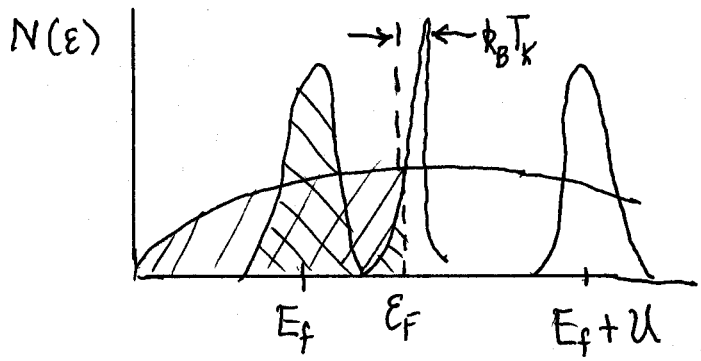
At high temperature the scattering becomes quasielastic (dashed line):
$\chi''(E) = \Gamma E / (E^2 + \Gamma^2)$
Crossover to
"relaxational spin dynamics"
$\chi(t) \sim e^{-\Gamma t}$



Murani, PRB 50 (1994) 9882

Anderson Impurity Model (AIM)

$$
\begin{aligned}
H_{AI} &= H_0 + H(i) + V(i) \\
H_0 &= \sum_k \varepsilon_k \mathbf{n}_k \\
H_f(i) &= E_f\ \mathbf{n}(i) + U\ \mathbf{n}_{f\uparrow}(i)\ \mathbf{n}_{f\downarrow}(i) \\
V(i) &= \sum_k V_{kf}\ \mathbf{c}_k^+\ \mathbf{f}(i) + c.c.
\end{aligned}
$$



Although intended for dilute alloys, (e.g. $La_{1-x}Ce_x$) because the spin fluctuations are local, the AIM describes much of the physics of periodic IV compounds. It includes the basic physics of:

Highly localized 4f orbital at energy **E_f**
Hybridization with conduction electrons with strength **V**
Strong on-site Coulomb interaction **U** preventing other 4f occupancies

Characteristic features:

Kondo Resonance

a low energy peak in the renormalized density-of-states (DOS) at
$k_B T_K \sim \varepsilon_F \exp\{-E_f/(N_J V^2\ N(\varepsilon_F))\}$

Spin/valence fluctuation: localized, with ***characteristic energy*** $k_B T_K$

Crossover: to local moment behavior for $T > T_K$

Universality: Properties scale as T/T_K, $E/k_B T_K$, $\mu_B H/k_B T_K$

Basic predictions of the AIM

High temperature limit: Local Moment Paramagnet

Integral valence: $n_f \rightarrow 1$ $z = 2+n_f = 3$ Yb $4f^{13}(5d6s)^3$

Curie Law: $T\chi/C_J \rightarrow 1$ $C_J = N\, g^2\, \mu_B^2\, J(J+1)/\, 3\, k_B$

Full moment entropy: $S \rightarrow R\, ln(2J+1)$ $J = 7/2$ (Yb)

Quasielastic spin fluctuations: $\chi'' \sim \chi\,(T)\, E\, \Gamma/(E^2 + \Gamma^2)$

$\Downarrow$

CROSSOVER at Characteristic temperature T_K

$\Downarrow$

Low temperature limit: Fermi Liquid

Nonintegral valence ($n_f < 1$)

Yb $4f^{14-n}f\,(5d6s)^{2+n}f$

Pauli paramagnet:

$\chi(0) \sim n_f C_J/T_K$

Fermi liquid entropy

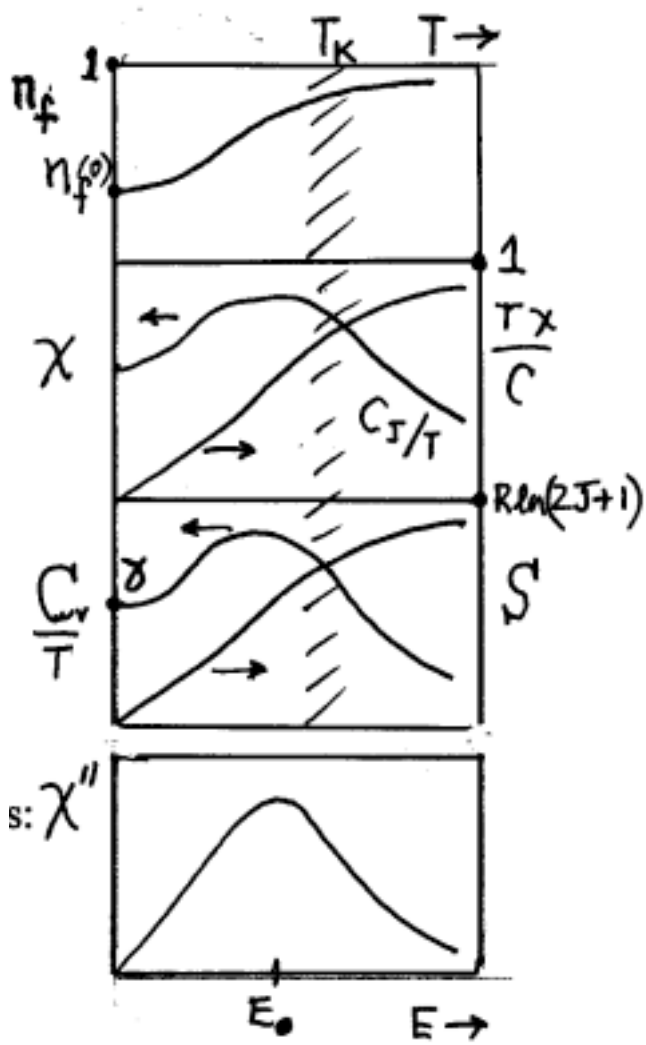
$S \sim C_v \sim \gamma\, T$

$\gamma = (\pi^2 k_B/3)\, (2J/2J+1)\, (n_f/T_K)$

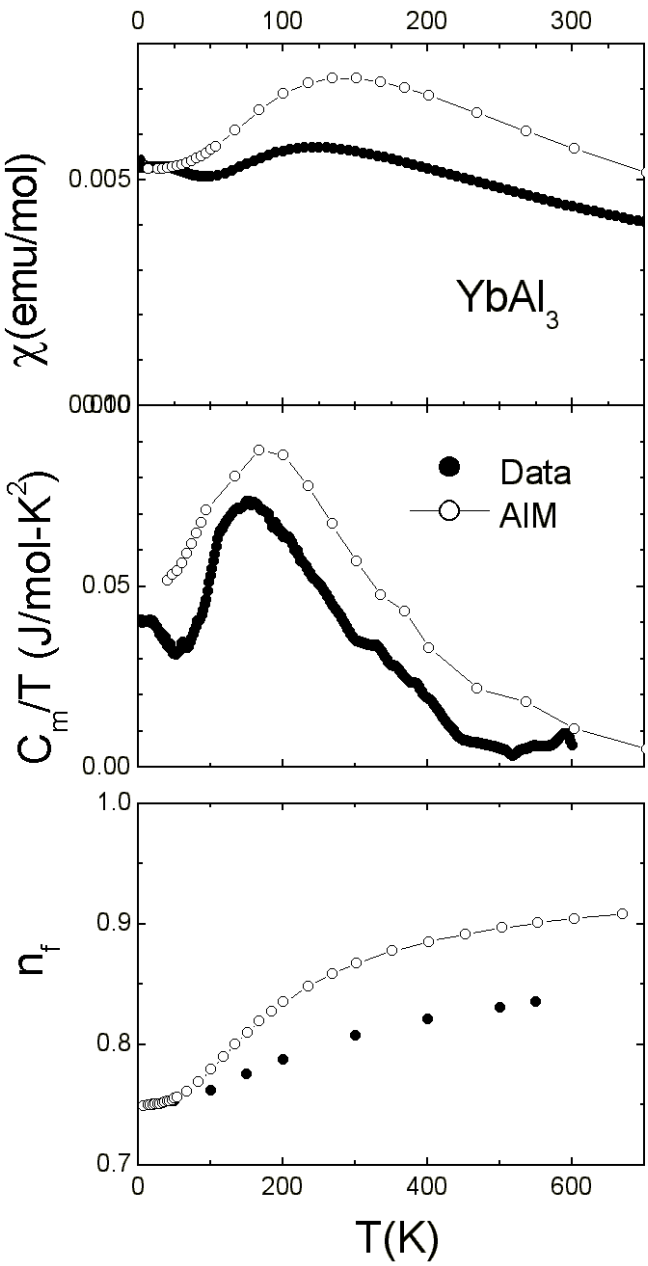
Damped inelastic spin fluctuations:

$\chi'' \sim \chi\,(T)\, E\, \Gamma/((E-E_0)^2 + \Gamma^2)$

$E_0 = k_B T_K$



$YbAl_3$: Susceptibilty, Specific Heat, 4f occupation Data vs. AIM



AIM parameters

(Chosen to fit $\chi(0)$, $n_f(0)$ and $\gamma(LuAl_3)$)

W = 4.33eV
E_f = -0.58264eV
V = 0.3425eV
T_K = 670K

Lawrence, Ebihara, Cornelius et al
e.g. PRL88 (2002) 117201

Comparison to AIM (continued)

Neutron scattering:

For the low-T Lorentzian power function, experiment gives

$E_0 = 40\text{meV}$ and $\Gamma = 25\text{meV}$

while the AIM calculation gives

$E_0 = 40\text{meV}$ and $\Gamma = 22\text{meV}$

The experiment also exhibits a crossover to quasielastic behavior that is expected in the AIM

Wilson ratio:

The AIM predicts that the normalized ratio of susceptibility to specific heat should be

$$(\pi^2 R/3C_J)\chi(0)/\gamma \cong 1 + (1/2J) = 8/7 = 1.14$$

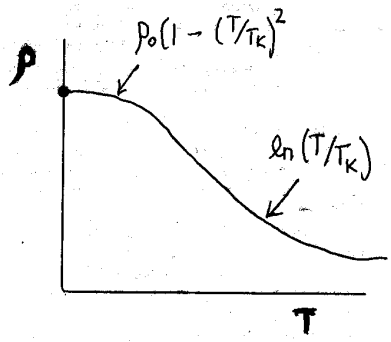
The experiment gives 1.3-1.4.

Overall agreement:

The AIM, with parameters chosen to fit $\chi(0)$ and $n_f(0)$
does an excellent job of fitting the neutron spectral parameters and fits the specific heat coefficient to within 20%.
It predicts the temperatures T_{max} of the maxima in susceptibility and specific heat to within 20%.
But the AIM predictions evolve more slowly with temperature than the data
and there are low temperature anomalies.

TRANSPORT BEHAVIOR OF IV COMPOUNDS

The AIM predicts a finite resistivity at T = 0 due to unitary scattering from the 4f impurity.



In an IV compound, where the 4f atoms form a periodic array, the resistivity must vanish.

(Bloch's law)

Typically in IV compounds

$\rho \sim A\,(T/T_0)^2$

This is a sign of Fermi Liquid "coherence" among the spin fluctuations.

$YbAl_3$ ρ vs T^2

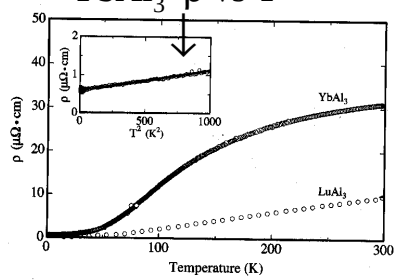


Fig. 1. Temperature dependence of the electrical resistivity of $YbAl_3$ and $LuAl_3$. The inset shows the T^2-dependence of the resistivity.

Ebihara et al
Physica B281&282 (2000) 754

In $YbAl_3$ the T^2 behavior of the resistivity is observed below 30K.

While the AIM is qualitatively good
(and sometimes quantitatively, e.g. $YbAgCu_4$)
for χ (T), C_v(T), n_f(T) and $\chi''(\omega;T)$
to get correct transport behavior and to determine the Fermi surface

Coherent Fermi Liquid behavior ⇒
Theory must treat 4f lattice

.

Two theoretical approaches to the Fermi Liquid State

Band theory: Itinerant 4f electrons

Calculate band structure in the LDA. Then, either

a) Add correlations through LDA + U, or

b) Add Kondo physics through Renormalized Band Method

Anderson Lattice Model: Localized 4f electrons

Various approximations, for example:

a) Ignore intersite contributions

(Yoshimori and Kasai, Journ. Mag. Mag. Mat. 31-34 (1983) 475)

b) Treat intersite contributions in NCA

(Georges et al, PRL 85 (2000) 1048)

c) Treat intersite contributions in Dynamic Mean Field Appx.

(Jarrell et al, PRB 55 (1997) R3332)

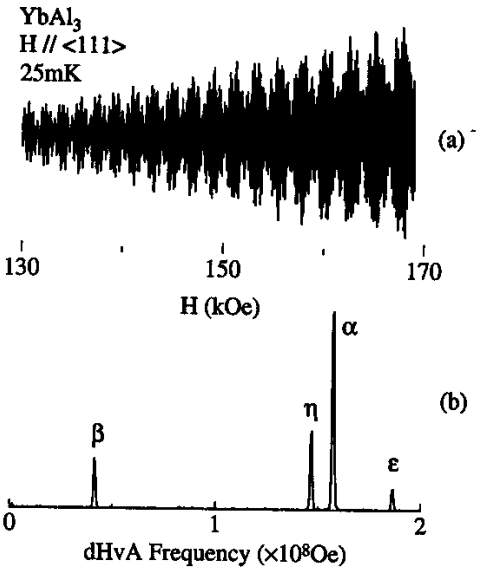
Bloch's law is satisfied for all approximations.

Approximations such as a) maintain single site thermodynamics

b) and c) give anomalies relative to single site thermodynamics

De Haas van Alphen and the Fermi surface

Figures from Ebihara et al, J Phys Soc Japan 69 (2000) 895



ig. 1. (a) DHvA oscillation and (b) its FFT spectrum for the apan field along ⟨111⟩ in $YbAl_3$.

The **de Haas van Alphen** experiment measures oscillations in the magnetization as a function of inverse magnetic field.

The frequency of the oscillations is determined by the areas S of the extremal cross sections of the Fermi surface in the direction perpendicular to the applied field.

$$M = A \cos(2\pi F/H)$$

$$F = (hc/2\,\pi e)\,S$$

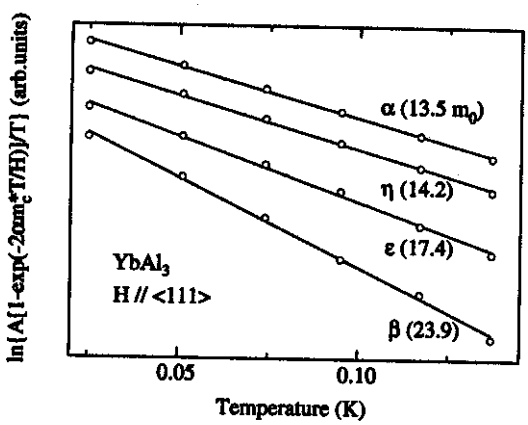


Fig. 3. Mass plot for the field along ⟨111⟩ in $YbAl_3$.

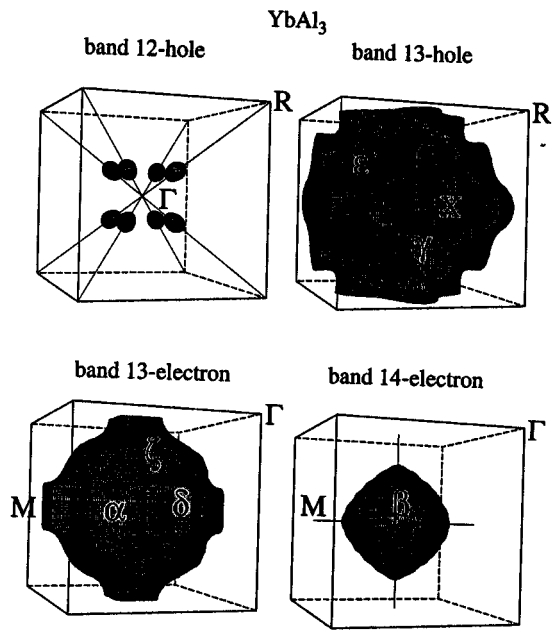


Fig. 7. Modified Fermi surfaces in $YbAl_3$. The band 13-hole and the band 13-electron Fermi surface represent the same Fermi surface.

The temperature dependence of the amplitude determines the effective mass m*

$$A = 1/\sinh(Qm^*T/H)$$

where Q is a constant

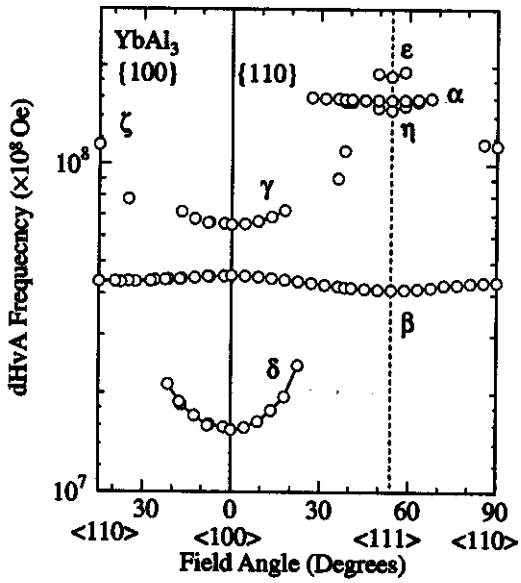
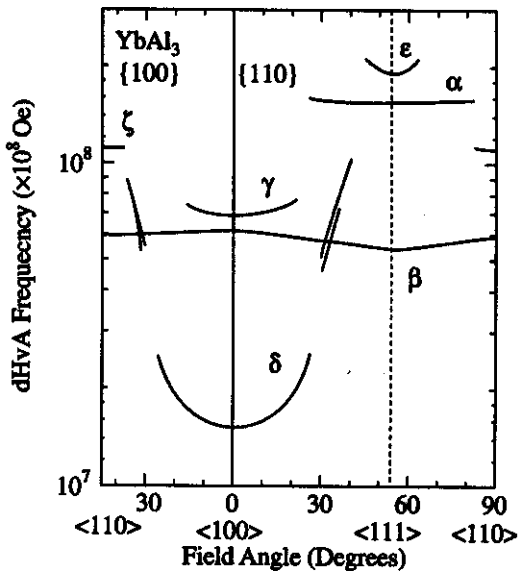


Fig. 2. Angular dependence of the dHvA frequency in $YbAl_3$.

EXPT



THEORY

For IV compounds LDA gives the correct extremal areas!

One-electron band theory (LDA) treats 4f electrons as itinerant.
Does a good job of treating the 4f-conduction electron hybridization.
Correctly predicts the topology of the Fermi surface seen by dHvA.

But: **LDA strongly underestimates the effective masses!**

LDA smooths the (very local!) Coulomb correlations.
This is a bad approximation for 4f electrons.
LDA badly overestimates the 4f band widths.
Consequently it strongly underestimates the effective masses:

LDA: $m^* \sim m_e$
dHvA: $m^* \sim 13\text{-}24\ m_e$

LDA alone makes $YbAl_3$ be divalent.
(The correct valence and the Fermi surface shown above was obtained by forcing the 4f level energy.)

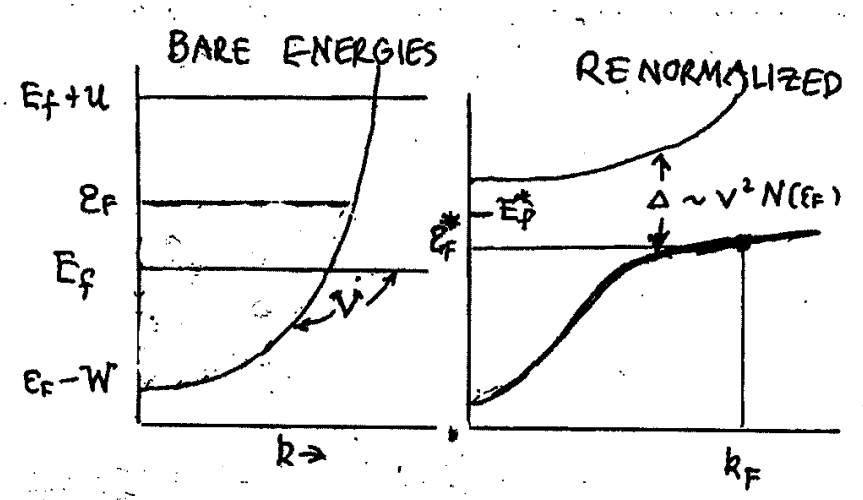
Large masses and intermediate valence also can be obtained by LDA + U

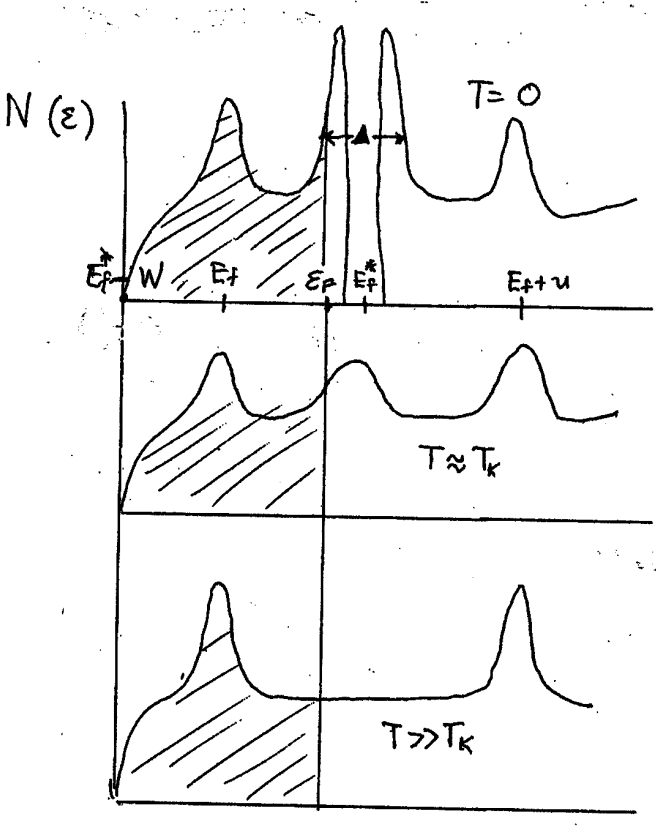
ANDERSON LATTICE

For a periodic IV compound there is one 4f on every site so the appropriate model is

$$H = \Sigma_k \varepsilon_k \mathbf{n}_k + \Sigma_i \{E_f \mathbf{n}_{fi} + U \mathbf{n}_{fi\uparrow} \mathbf{n}_{fi\downarrow} + \Sigma_k [V_{kf} \mathbf{c}^+_k \mathbf{f}_i + cc]\}$$

This leads to a coherent band structure with (renormalized) hybridized bands near the Fermi energy. The bands exhibit a hybridization gap; the Fermi level lies in the high DOS region due to the large admixture of 4f states. The large DOS is responsible for the large m*.





The structure renormalizes away with increasing temperature:

For very low $T << T_K$
Fully hybridized bands

For $T \sim T_K$,
No gap
Incoherent Kondo resonances

For $T >> T_K$
Local moments uncoupled from band electrons

Optical conductivity

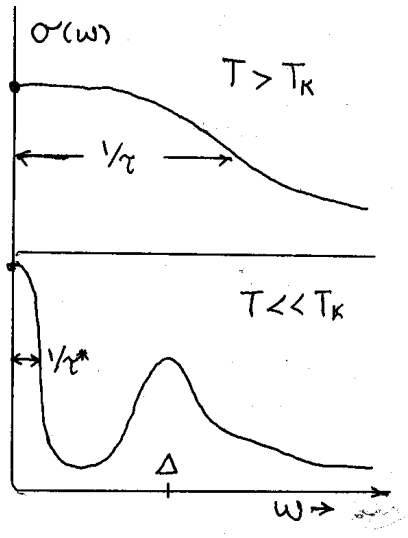
BEST EVIDENCE FOR THE HYBRIDIZATION GAP AND ITS RENORMALIZATION WITH TEMPERATURE

High temperature:

Normal Drude behavior:

$\sigma'(\omega) = (ne^2/m_b)\{\tau / (1 + \tau^2\omega^2)\}$

m_b: bare band mass τ : relaxation time.



⇓

CROSSOVER

⇓

Low temperature:

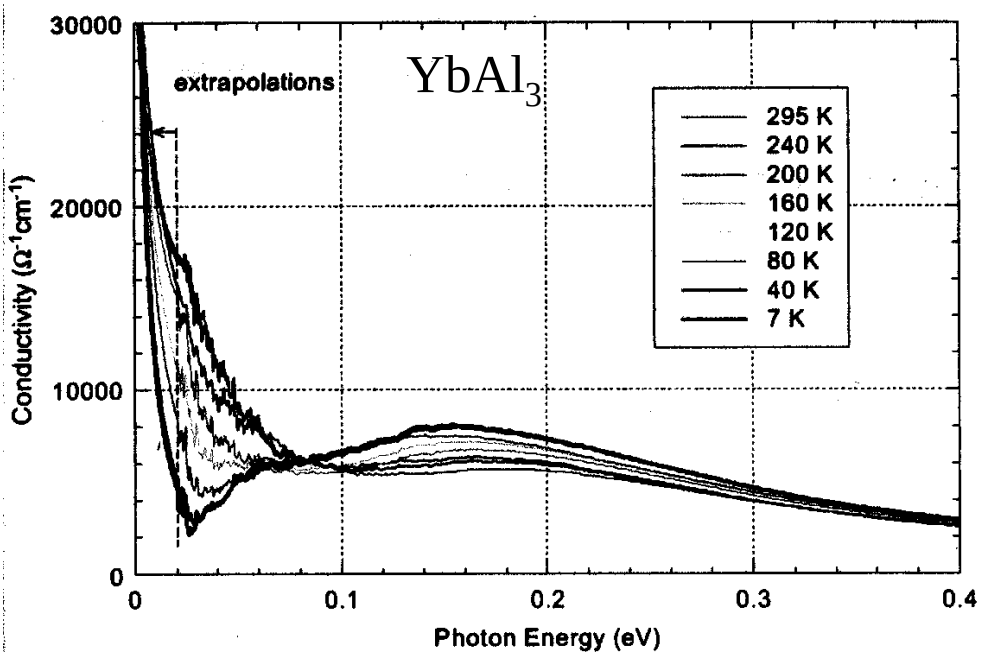
1) Infrared absorption peak from transitions across hybridization gap

2) *Very* narrow Drude peak. Both m and τ renormalized.

$m_b \rightarrow \lambda\, m_b = m^*$ $\tau \rightarrow \lambda\, \tau = \tau^*$

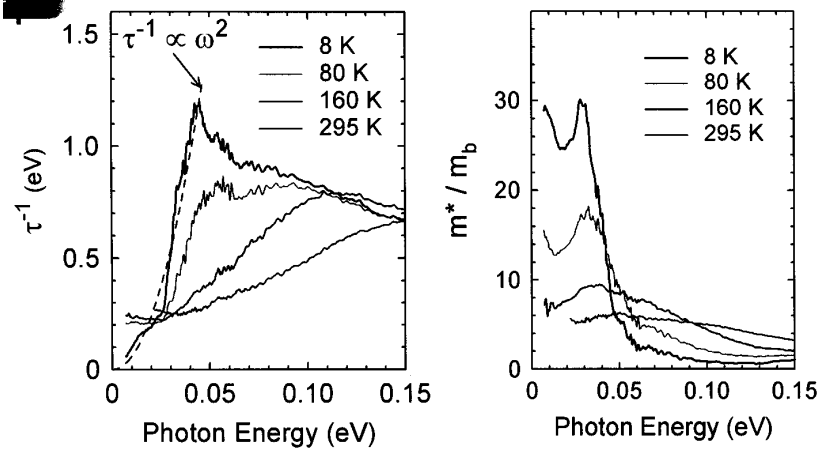
$\sigma'(\omega) = ne^2\,[\tau^*/m^*]\,\{1/(1 + \tau^{*2}\omega^2)\}$

Okamura, Ebihara and Namba, unpublished



Note: **IR conductivity invariant with T below 40K**

Extended Drude analyses



- Fermi liquid behavior ($1/\tau \sim \omega^2$)
- Mass enhancement (m^*/m_b =25-30)
  → Heavy-mass Fermi liquid

Assuming frequency-dependent scattering
$\sigma(\omega) = (ne^2/m_b)\,[\gamma(\omega) - i\,\omega]^{-1}$
then the mass enhancement
$m^* = \lambda m_b$
$\lambda(\omega) = -\mathrm{Im}[\gamma(\omega)]/\,\omega$
is both frequency and temperature dependent

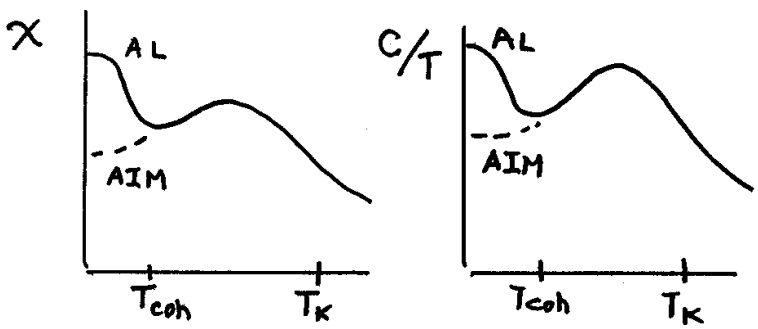
For $YbAl_3$ this procedure gives m*~25-30, comparable to the dHvA masses.

Two energy scales and slow crossover in the Anderson Lattice

While the transport behavior and the Fermi surface (dHvA) are affected by Fermi liquid coherence, we have seen that experimental quantities such as the specific heat, susceptibility, valence and spin dynamics are qualitatively in good accord with the predictions of the AIM over a broad range of temperature. This reflects highly localized spin/valence fluctuations

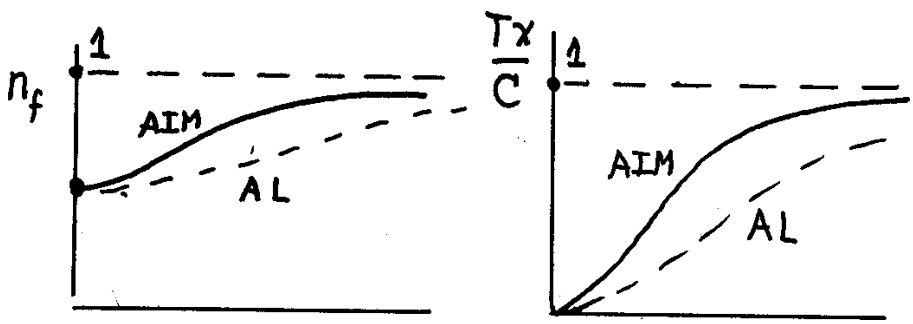
Nevertheless, recent theory for the Anderson Lattice suggests that the behavior of these quantities can differ in two ways from the predictions of the AIM:

1) Non-universal low temperature scale for coherence Low temperature anomalies



Antoine Georges et al,
PRL 85 (2000) 1048

2) Slow crossover from Fermi Liquid to Local Moment



Mark Jarrell et al
PRB55 (1997) R3332

Theory predicts that these differences become magnified when the conduction electron density is low.

Slow crossover in $YbAl_3$

Slow crossover has been reported for $\chi(T)$ and $n_f(T)$ for $YbXCu_4$

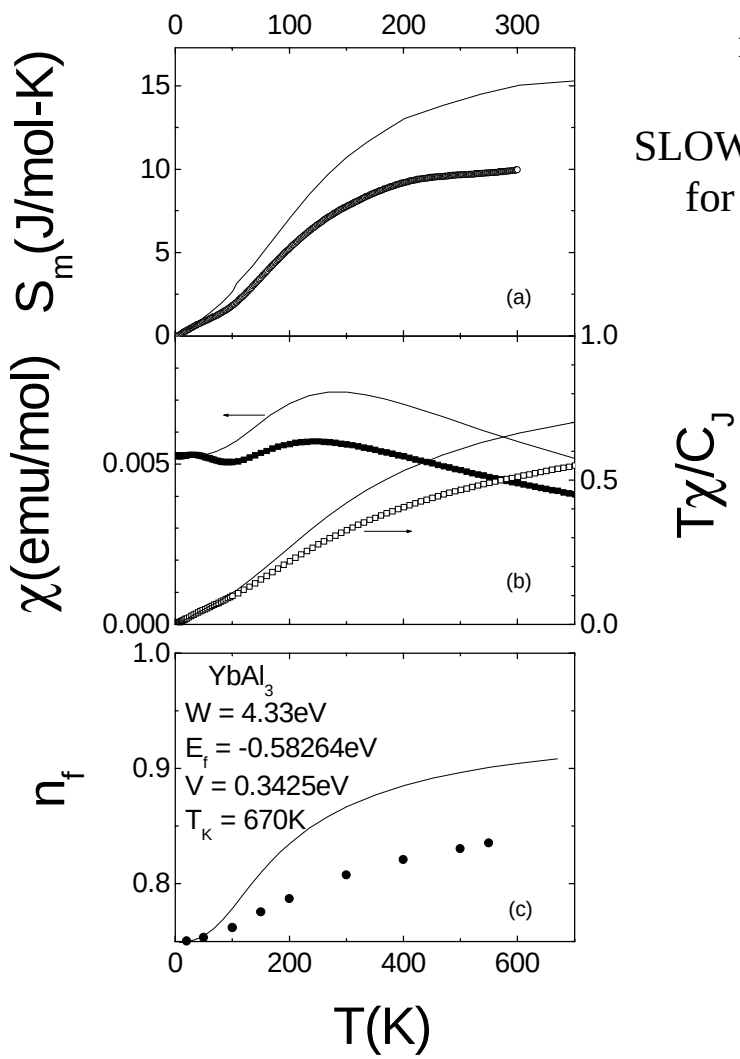
(Lawrence et al, PRB 63 (2001) 054427)

and correlated to electron density determined from Hall coefficient

$$n_e = 1/R_H e$$

$YbAgCu_4$, $YbTlCu_4$ $n_e > 1$e/atom No slow crossover

$YbMgCu_4$, $YbZnCu_4$ $n_e < 1$e/atom Slow crossover



For $YbAl_3$

$n_e = 1/R_H e = 0.5$ e/atom

and

SLOW CROSSOVER OBSERVED

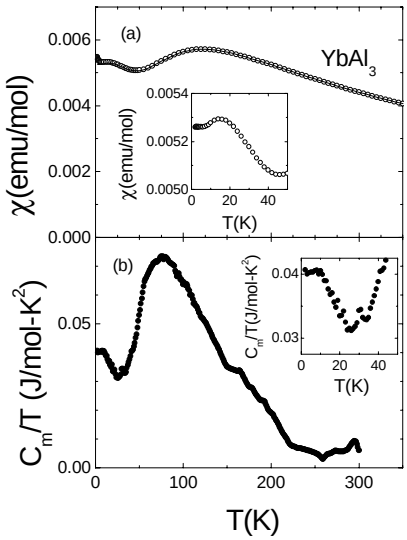
for entropy, susceptibility and

4f occupation number

Symbols: expt. Data

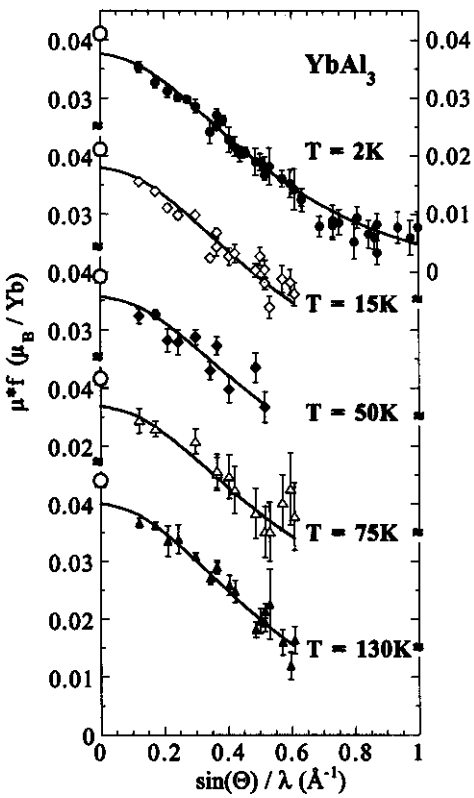
Lines: AIM

Low temperature anomalies in $YbAl_3$



Cornelius et al
PRL 88 (2002) 117201

Above 40K the **susceptibility** and **specific heat** correspond qualitatively to the predictions of the AIM. Below 30-50K, **anomalies** are observed.



Hiess et al
J Phys: Cond. Mat 12 (2000) 829

No form factor anomaly:
The neutron form factor measures the spatial distribution of magnetization around the Yb site.

At most temperatures the form factor has the same Q (or r) dependence $f^2(4f;Q)$ as the 4f radial function.

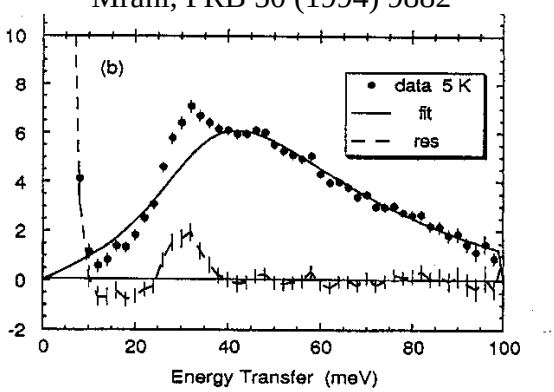
In $CePd_3$ and $CeSn_3$ at low T a more diffuse 5d component $f^2(5d;Q)$ occurs:
$f^2(Q) = a^2\, f^2(4f) + (1-a^2)\, f^2(5d)$
This 5d contribution gives rise to an anomaly in the low temperature susceptibility similar to that of $YbAl_3$.

However, in $YbAl_3$, there is ***no form factor anomaly*** -- the magnetization density is that of the 4f orbital at all temperatures (solid lines).

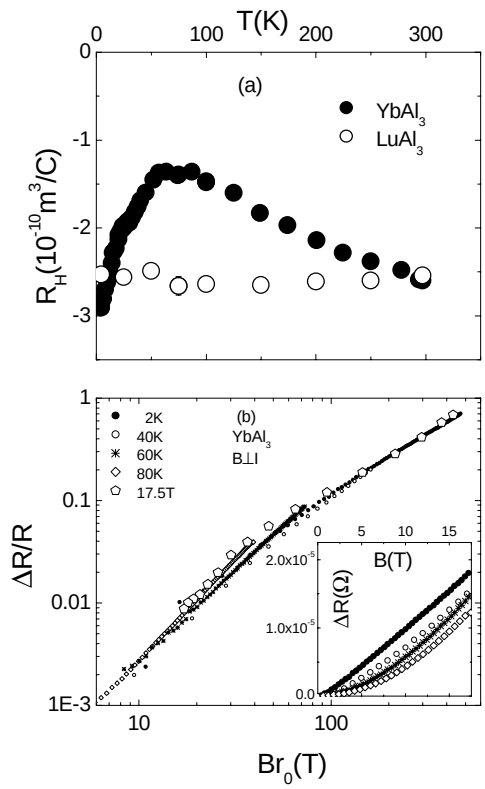
New peak in low temperature spin dynamics

We have seen that for most temperatures and energies the magnetic neutron scattering in $YbAl_3$ follows the predictions of the AIM, with a Lorentzian power spectrum with $E_0 = 40meV$ and $\Gamma = 25meV$

Mrani, PRB 50 (1994) 9882



At low T, <u>there is an additional narrow peak</u> with
$E_0 = 30meV$ and $\Gamma = 5meV$
This peak vanishes above 50K, and hence appears to be a property of the fully coherent ground state.



Magnetotransport anomalies

Anomalies in the Hall coefficient and magnetoresistance are observed in this same temperature range.

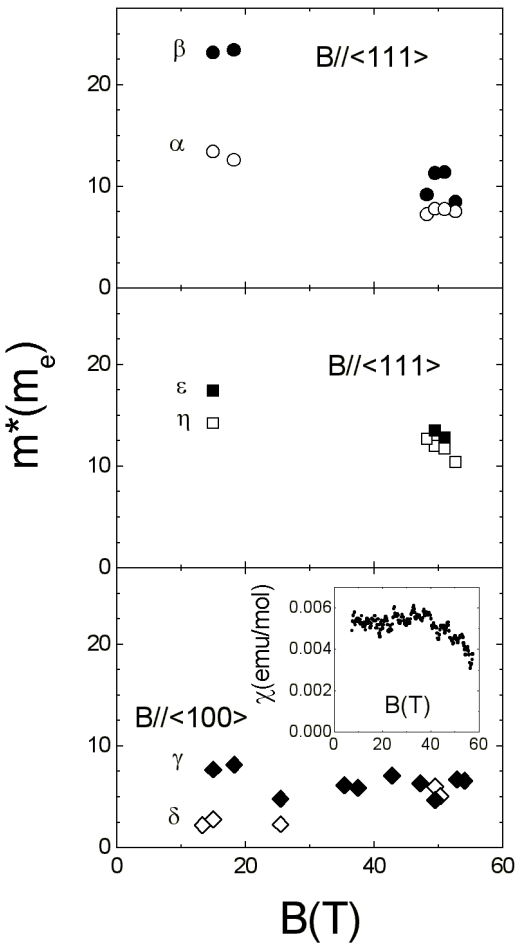
Since $R_H \sim 1/ne$ this suggests a change in carrier density. The onset of coherent Fermi liquid behavior appears to involve a change in the Fermi surface.

Cornelius et al
PRL 88 (2002) 117201

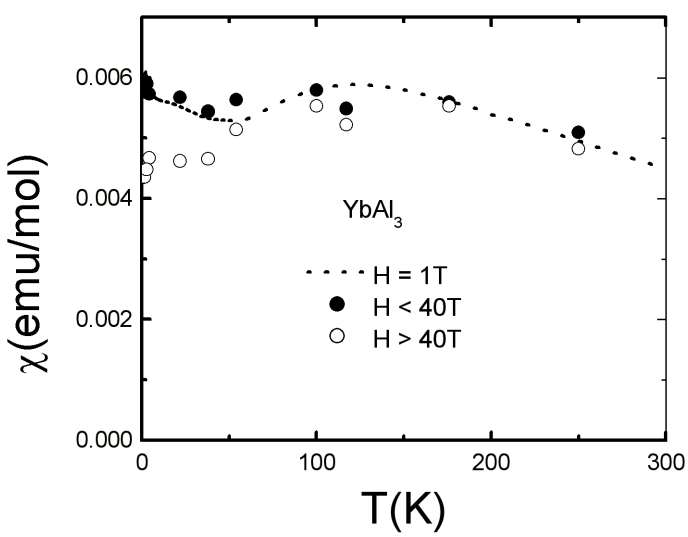
Field dependent masses in $YbAl_3$

Ebihara, Cornelius, Lawrence, Uji and Harrison

cond-mat/0209303



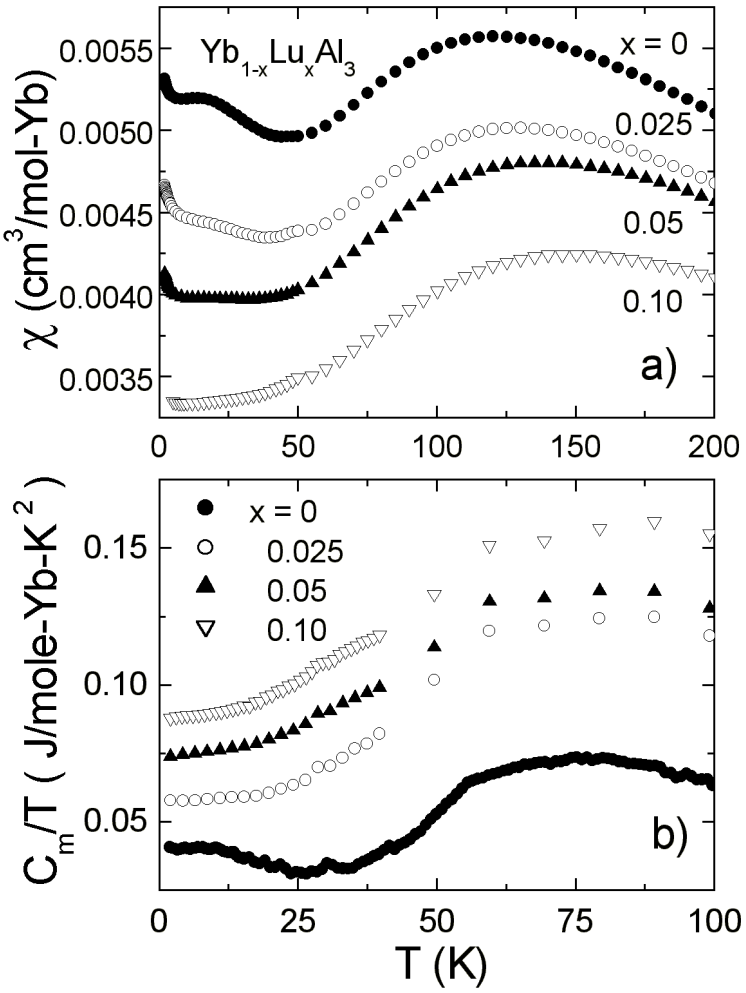
High field dHvA shows that the effective masses for H//<111> decrease substantially for H > 40T. This field is much smaller than the Kondo field $B_K = kT_K/gJ\mu_B$ required to polarize the f electrons, but is of order k_BT_{coh}/μ_B.



A field of this magnitude also suppresses the low temperature susceptibility anomaly.

It is as though that the system exhibits a crossover from a anomalous high mass Fermi Liquid state to a non-anomalous moderately enhanced Fermi liquid state for $\mu_BH > k_BT_{coh}$.

EFFECT OF DISORDER ON THE LOW TEMPERATURE ANOMALIES



The low temperature anomalies in the susceptibility and specific heat are very sensitive to alloy disorder, and disappear for alloy concentrations as small as x = 0.05 in $Yb_{1-x}Lu_xAl_3$. Apparently the enhanced masses observed below T_{coh} are very sensitive to lattice coherence.

Low temperature anomalies and the onset of fully coherent Fermi liquid behavior in $YbAl_3$

In $YbAl_3$, the Kondo temperature $T_K = 670K$ sets the main scale for the crossover from local moment behavior to nonmagnetic behavior. The behavior is that of uncorrelated Kondo impurities.

Below a lower temperature scale for the onset of coherence

$$(T_{coh} \sim 30\text{-}50K \sim n_f T_K / (2J+1))$$

the following occurs:

T^2 behavior of the resistivity

Fully renormalized hybridization gap in optical conductivity
(no change in mid-IR conductivity below 40K)

Fully renormalized optical mass in optical conductivity

Large effective masses in dHvA

In addition, **anomalies** appear in the following quantities:

Hall effect (and magnetoresistance)

Susceptibility

Specific heat coefficient C/T

New peak (in addition to Kondo peak) in the spin dynamics

The large dHvA masses and the susceptibility anomaly are suppressed by a magnetic field of 40T. This field is much smaller than the Kondo field $B_K = k_B T_K / g J \mu_B$ required to polarize the f-electrons but is of order $k_B T_{coh}$.

The susceptibility and specific heat anomalies are extremely sensitive to alloy disorder, suggesting that the enhanced effective masses are very sensitive to lattice coherence.

Plus: slow crossover from Fermi liquid to local moment regime.