Optics and transport in paramagnetic heavy Fermions

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The periodic Anderson model:

- Basic model for lanthanide-based heavy electron materials

  ---- heavy fermion (**HF**) and intermediate valence (**IV**) metals, and Kondo insulators (**KI**).

- 'Simple'/minimalist: a single correlated f-level in each unit cell hybridizes locally to an uncorrelated conduction band.

- So to what extent are real materials captured by the PAM?

  Clearly requires explicit comparison to experiment.

  A central aim here: focus on *d.c. transport* + *optical properties* of 'normal' paramagnetic phase.

- Basic framework: Dynamical Mean-Field Theory (**DMFT**).

- Within DMFT framework, require theory that can:-

  --- (a) can handle strong correlations (and weak),

  (b) on *all* energy and/or temperature scales relevant to experiment

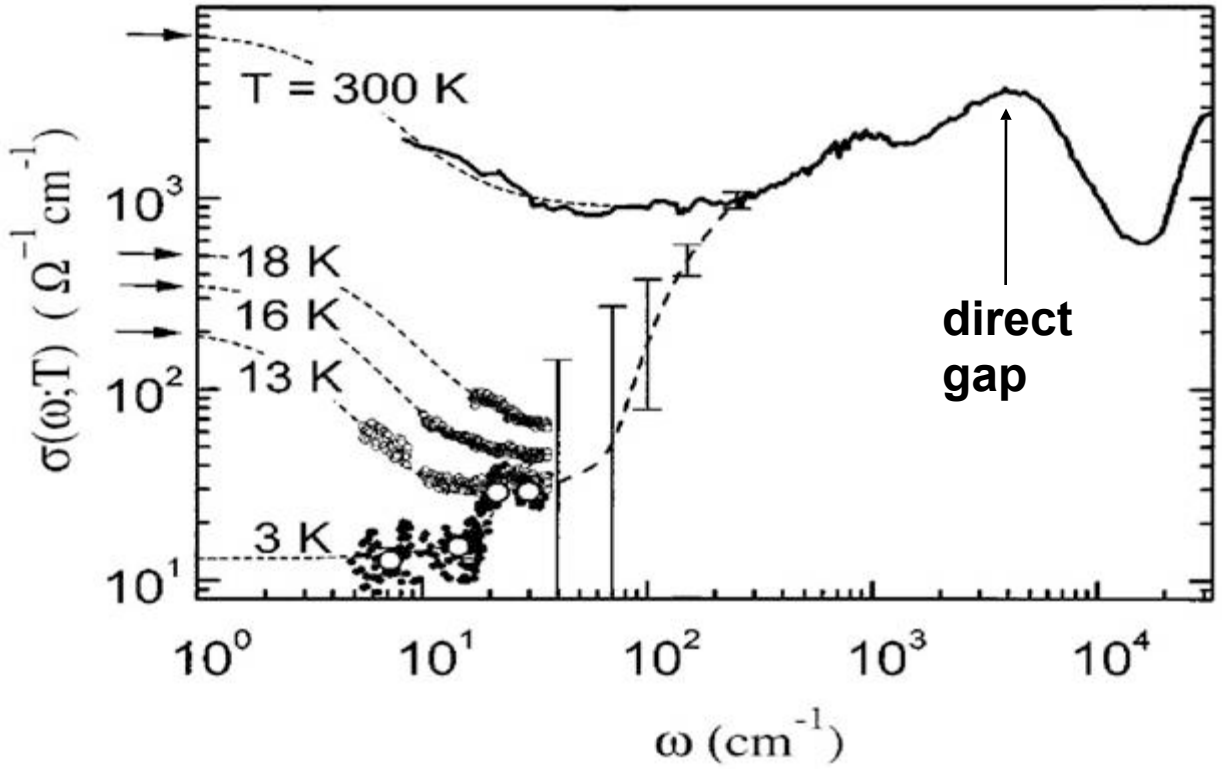
  (not just e.g. low-energy dictates of asymptotic Fermi liquid theory).

To that end employ here the local moment approach (**LMA**); meets above criteria.

Experimental example:

Optical conductivity, $\sigma(\omega;T)$ vs ω, and its thermal evolution.

Kondo insulator SmB_6:



- Rich frequency dependence, from microwave to NIR.
- Strong thermal evolution at low-T.

Gorshunov et al Phys Rev B **59,** 18081 (1999).

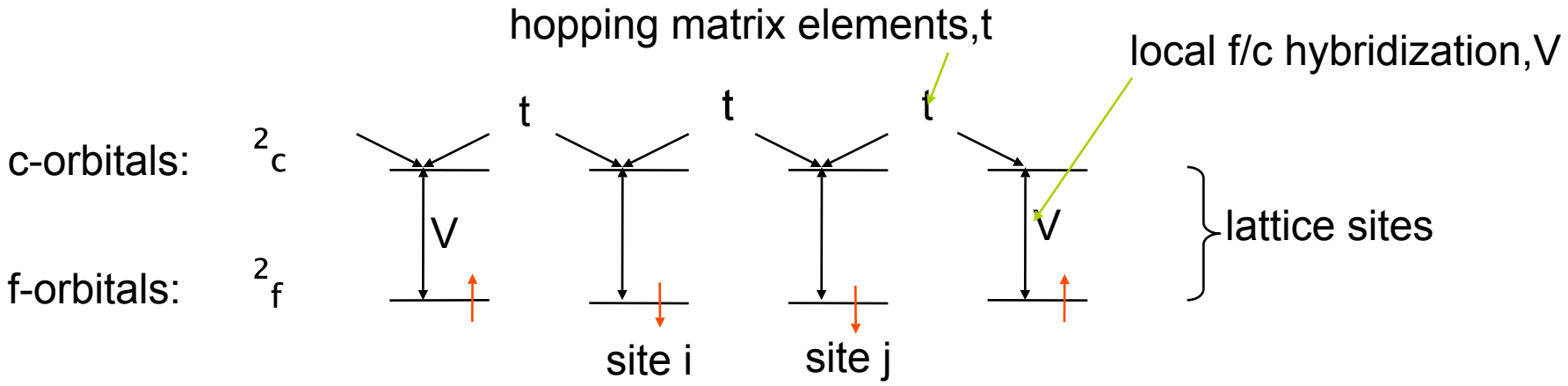
Nanba et al, Physica B, **186-188**, 440 (1993)

Outline:

- Model, briefly.
- Naive bandstructure picture.
- Theory: DMFT + LMA, a sketch.
- Theoretical results for transport/optics.
  --- primary focus on strongly correlated regime, and scaling.
- Explicit comparison to experiment:
  SmB_6 --- a strongly correlated Kondo insulator.
  CeB_6 --- a strongly correlated heavy fermion metal.
  $YbAl_3$ --- an intermediate valence metal.
  $CeAl_3$ --- a classic HF (+ 'odd' optics).

Periodic Anderson Model (PAM)

Each lattice site has two orbitals, 'c' and 'f'; schematically:



$$\hat{H} = \hat{H}_{cond} + \hat{H}_f + \hat{H}_{hybridn}$$

$$\hat{H}_{cond} = \sum_{i,\sigma} {}^2_c c^{\dagger}_{i\sigma} c_{i\sigma} \; i \; t \sum_{(i,j),\sigma} c^{\dagger}_{i\sigma} c_{j\sigma} \; ' \sum_{k,\sigma} ({}^2_c + {}^2_k) c^{\dagger}_{k\sigma} c_{k\sigma}$$

$$\hat{H}_{hybridn} = V \sum_{i,\sigma} (f^{\dagger}_{i\sigma} c_{i\sigma} + c^{\dagger}_{i\sigma} f_{i\sigma})$$

Plus the correlated f-level Hamiltonian:

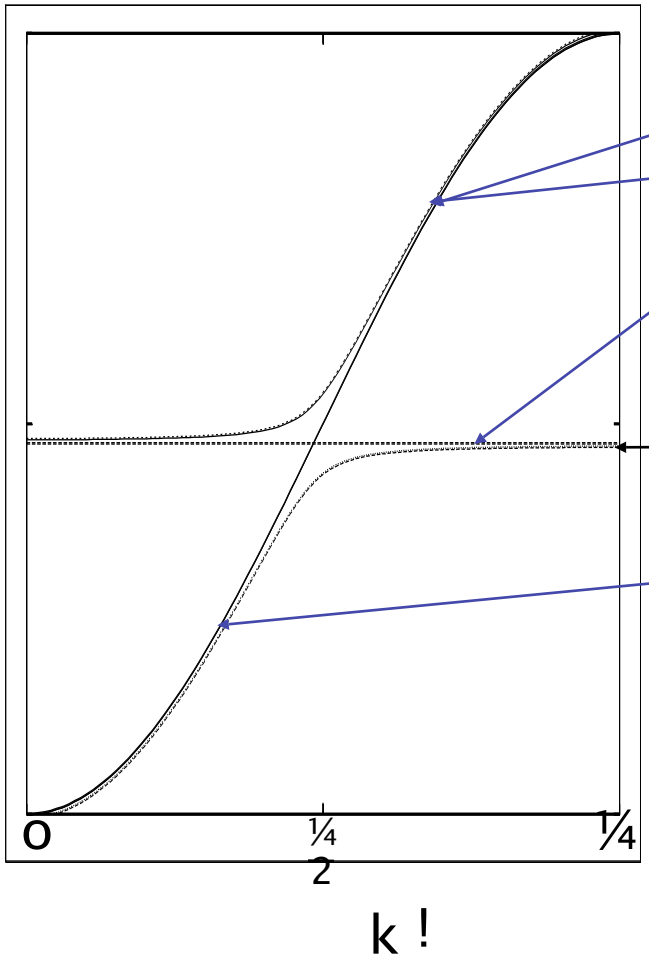
$$\hat{H}_f = \sum_i [{}^2_f \hat{n}_i + U \hat{n}_{i"} \hat{n}_{i\#}]$$ …… with local coulomb repulsion U.

Four 'bare' parameters: ^2_c; V; U; and ^2_f:

A first stab at the problem: effectively neglect interactions, i.e.

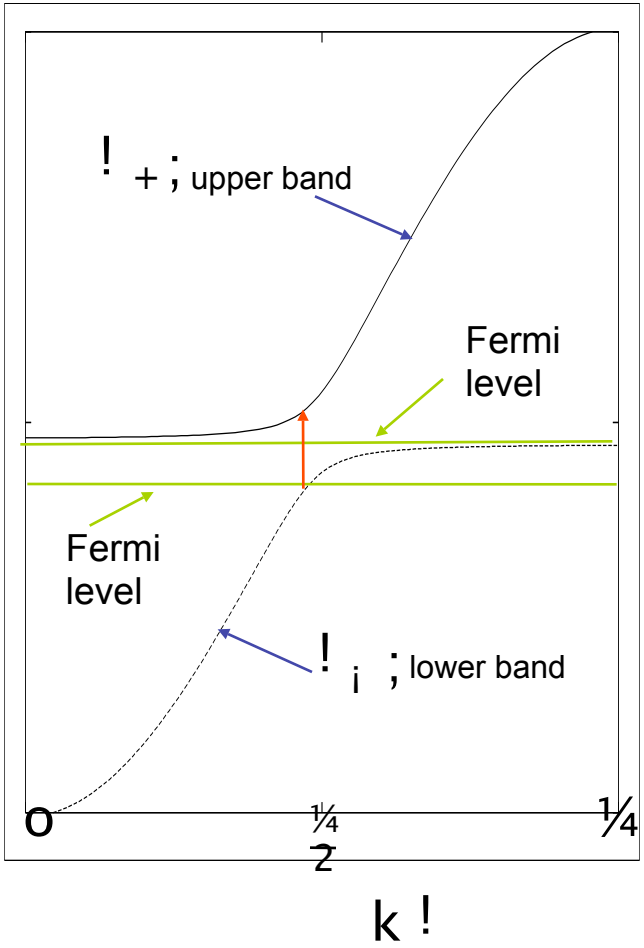
Bandstructure view .....

[Either 'straight' or 'renormalized' bandstructure.]



Way too naive.....

Bandstructure view.....



• If Fermi level lies in gap system a 'hybridization gap' insulator; conductivity ¾(T) = 0

… for **all** T!

• Generically, system metallic, conductivity

¾(T) = 1 (½(T) = 0)

… for **all** T!

• The lowest energy optical excitation is the direct gap ( ), ¢ dir » 2V: no optical excitations whatever below this relatively high energy.

Again, qualitatively wrong.

• All due of course to complete absence of e-e scattering inherent in any effective 1-e approach.

Background theory: DMFT and LMA ---- sketch.

Focus on the *local* c- and f-electron propagators, $G^c(!) \equiv G^c_{ii}(!)$ (and $G^f(!) \equiv G^f_{ii}(!)$). Sufficient.

A. Free conduction band, V=0 ---- no coupling to f-levels. Trivial: tight-binding band.

local c-propagator: $g^c(!) = \frac{1}{N}\sum_k g^c(²_k;!) = \frac{1}{N}\sum_k \frac{1}{[! + i\ ²_c i\ ²_k]}$ (1)

$\Rightarrow$ $g^c(!) = \int_{-1}^{1} d² \frac{½_0(²)}{! + i\ ²_c i\ ²}$ (2a) $(½_0(²) = \frac{1}{N}\sum_k \pm(² i\ ²_k)$ --- free DoS)

$= H(! + i\ ²_c)$ (2b) ---- Hilbert transform.

This is 'k-space view' of *local* propagator. What about a '*local view*' ---e.g. re locator expansion? Define Feenberg self-energy by:

$g^c(!) = \frac{1}{! + i\ ²_c i\ S_0(!)}$ (2c)

(2b,c) $\Rightarrow$ $g^c = H(\frac{1}{g^c} + S_0)$ i.e.

$S_0(!) \equiv S[g^c]$ (3) -- a functional of g^c

B. $V \neq 0$ --- **coupled to correlated f-levels.**

local c-propagator: $$G^c(\omega) = \frac{1}{\zeta(\omega) - \Sigma} \qquad (4a)$$

with $$\zeta(\omega) = \omega + i\eta - \varepsilon_c - \frac{V^2}{\omega + i\eta - \varepsilon_f - \Sigma_f(\omega)} \qquad (4b)$$ ($\Sigma_f(\omega)$ local --- DMFT)

and (key): $$\Sigma \equiv \Sigma[G^c] \qquad (4c)$$

---- the *same* functional of G^c as of g^c when V=0.

⇒ $$G^c(\omega) = H(\zeta) = \int_{-1}^{1} d\varepsilon \frac{\rho_0(\varepsilon)}{\zeta(\omega) - \varepsilon} \qquad (5)$$

So procedure clear (in principle):

----- *given* f-electron self-energy $\Sigma_f(\omega)$, (4b) gives $\zeta(\omega)$.

----- (5) then gives $G^c(\omega) = H(\zeta)$ as sought.

[and the f-level propagator follows directly from G^c].

BUT: ……..

.....the hard part is of course $\Sigma_f(!)$.

- Perturbation theory (PT) in U fine in principle:

  --- but can't handle strong interactions in practice.

- Diagrammatic PT, based on usual Hartree M-F propagators, decomposes $\Sigma_f(!)$ as:

$$\Sigma_f(!) = \text{(loop)} + \Sigma_f^0(!)$$

$$= \frac{U}{2} n_f + \Sigma_f^0(!)$$

Hartree M-F

dynamics constructed from Hartree propagators

But: the 'natural' diagram resummations here suffer from well known divergences/ non-analyticities.

Why? Because procedure amounts to expansion about *Hartree* M-F saddle point. And ***that saddle point is generally unstable to local moment condensation***.

- The correct M-F saddle point then corresponds to *Hartree-Fock*, with local moment condensation s.t. $^1 = +|^1|$ or $-|^1|$ for each site --- the saddle point is locally doubly degenerate: $\mathcal{R}=A$ or B with $^1A = +|^1|$ and $^1B = -|^1|$

Should thus construct f-electron self-energies about *these* saddle points:

$$\tilde{\Sigma}_{\sigma}(\omega) = \quad + \quad \Sigma_{\sigma}(\omega) \qquad \sigma = A \text{ or } B$$

(&spin $\uparrow$ or $\downarrow$)

$$= \frac{U}{2}[n_f \; i \; \uparrow \; \sigma] + \Sigma_{\sigma}(\omega)$$

Hartree-*Fock* M-F: $^{1}_{A} = +j^{1}\,j\,.$
$^{1}_{B} = i\; j^{1}\,j\,.$

Dynamics post-MF, constructed from Hartree-*Fock* propagators.

And because saddle point degenerate, local c-propagator is

$$G^{c}(\omega) = \frac{1}{2}[G^{c}_{A\uparrow}(\omega) + G^{c}_{B\uparrow}(\omega)] \qquad (6a)$$

← rotationally invariant (because $G^{c}_{A\uparrow}(\omega) = G^{c}_{B\downarrow}(\omega)$)

$$= \frac{1}{2}\left\{\frac{1}{\zeta_{A\uparrow} - S[G^{c}]} + \frac{1}{\zeta_{B\uparrow} - S[G^{c}]}\right\} \qquad (6b)$$

with $\zeta_{A\uparrow}(\omega) = \omega + i\,\Gamma_{c} - \dfrac{V^{2}}{\omega + i\,\Gamma_{f} - \tilde{\Sigma}_{A\uparrow}(\omega)}$ $\quad (= \zeta_{B\downarrow}(\omega))$ (6c)

Again: because saddle point degenerate, local c-propagator is

$$G^c(\omega) = \tfrac{1}{2}[G^c_{A\alpha}(\omega) + G^c_{B\alpha}(\omega)] \qquad (6a)$$

← rotationally invariant (because $G^c_{A\alpha}(\omega) = G^c_{B\alpha}(\omega)$)

$$= \tfrac{1}{2}\left\{\frac{1}{\omega - \Delta_{A\alpha} - \Sigma[G^c]} + \frac{1}{\omega - \Delta_{B\alpha} - \Sigma[G^c]}\right\} \qquad (6b)$$

with $$\Delta_{A\alpha}(\omega) = \omega + i\,\epsilon^2 c\, i\,\frac{V^2}{\omega + i\,\epsilon^2 f\, i\,\tilde{\Sigma}_{A\alpha}(\omega)} \quad (= \Delta_{B\alpha}(\omega)) \qquad (6c)$$

Comparison of (6b) with $G^c(\omega) = \frac{1}{\omega - \Delta(\omega) - \Sigma}$ ⇨

$$\Delta(\omega) = \tfrac{1}{2}[\Delta_{A\alpha}(\omega) + \Delta_{B\alpha}(\omega)] + \frac{[\frac{1}{2}(\Delta_{A\alpha}(\omega) - \Delta_{B\alpha}(\omega))]^2}{\Sigma - \frac{1}{2}[\Delta_{A\alpha}(\omega) + \Delta_{B\alpha}(\omega)]} \qquad (7a)$$

with $$\Sigma = \Delta - \frac{1}{H(\Delta)} \qquad (7b)$$

So procedure clear:

--- given f-electron self-energies $\tilde{\Sigma}_{\alpha}(\omega)$, (6c) gives $\Delta_{\alpha}(\omega)$,

--- (7) is closed: solution gives $G^c(\omega) = H(\Delta)$ as sought. [$G^f(\omega)$ follows from $G^c(\omega)$].

This ‘two self-energy’ description underlies the LMA. It is general.

• Recall:

$$\tilde{\Sigma}_{A\sigma}(\omega) = \frac{U}{2}[n_f - \sigma j^{-1} j] + \Sigma_{A\sigma}(\omega) = \tilde{\Sigma}_{B-\sigma}(\omega) \qquad (8) \qquad [\sigma = \uparrow \text{ for } \alpha = \#\text{spins}]$$

pure MF

dynamics post-MF

key in practice!

Saddle point(s) no longer unstable, so 'natural' diagrammatic resummations for $\Sigma_{R\sigma}(\omega)$ don't exhibit divergences/non-analyticities. Can use with impunity.

• Can also be shown --- *generally* --- that adiabatic continuity to non-interacting limit (Fermi liquid behaviour) ⇨

$$\tilde{\Sigma}_{A\sigma}(\omega = 0) = \tilde{\Sigma}_{B\sigma}(\omega = 0) \qquad (9) \qquad [\omega = 0 \equiv \text{Fermi level}]$$

--- clearly <u>not</u> satisfied at <u>pure</u> MF alone (from (8)), where saddle point locally doubly degenerate. Reflecting?

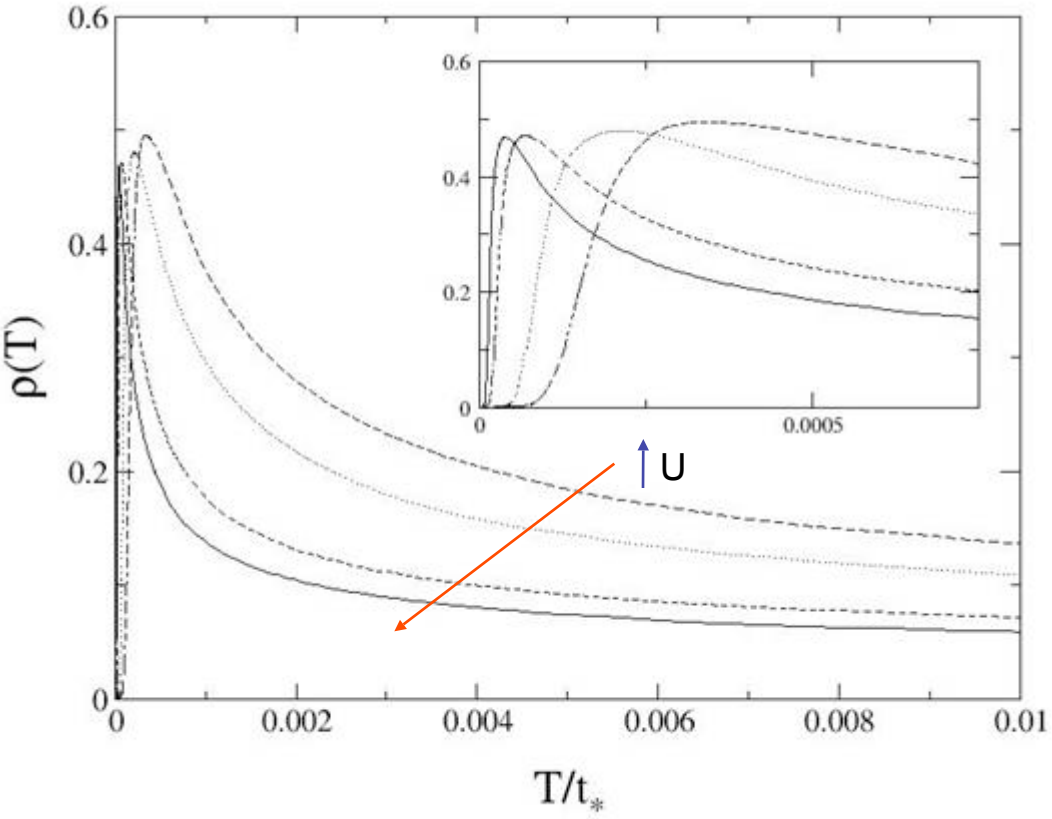
--- that (9) is/must be satisfied in general thus corresponds to **symmetry restoration**. Meaning?

[in practice, for specified $\{\Sigma_{R\sigma}(\omega)\}$, (9) self-consistently imposed.]

Comment: --- on the 'two-self-energy' description and NRG.

Briefly, **some results from the LMA**……(focus on strongly correlated HF regime).

: d.c. resistivity vs T, on an 'absolute scale' i.e. vs T/t_* where t_* sets the conduction electron bandwidth (on the eV scale).



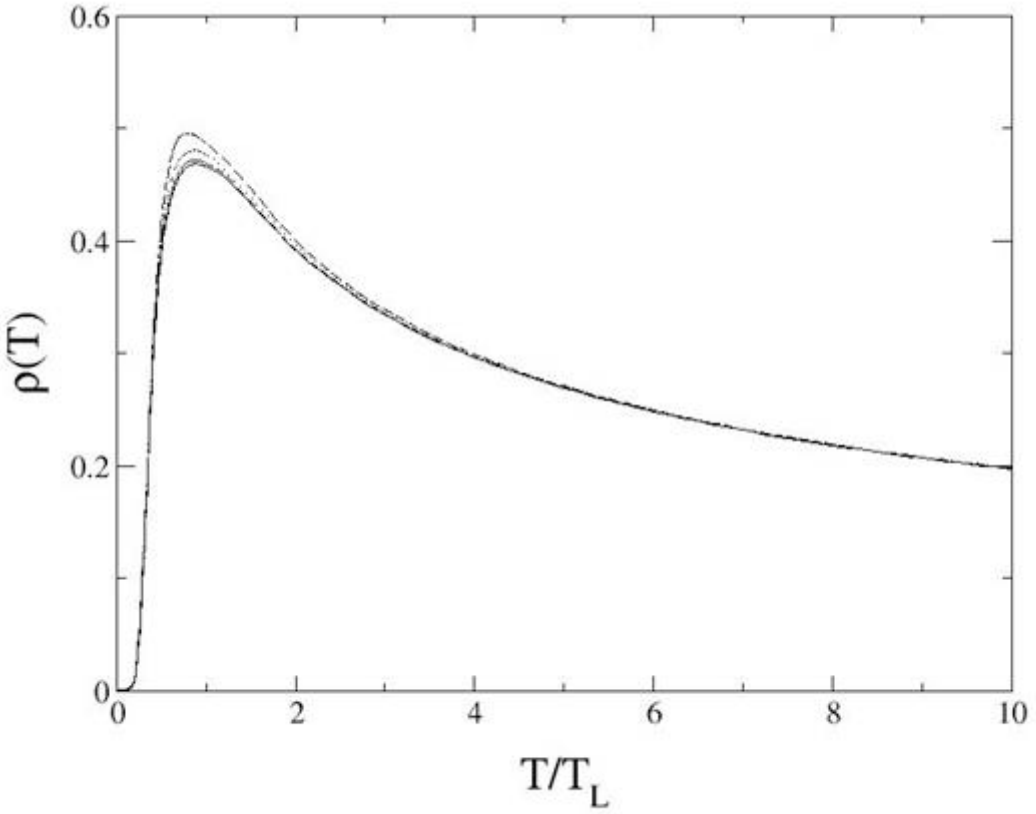
Shown for four increasing interaction strengths, $U/t_* = 4{:}6;\ 5{:}1;\ 6{:}1;\ 6{:}6{:}$

--- all v. different on this 'absolute' scale; attesting to exponential diminution of low-energy scale with increasing U/t_*:

• Now let's simply rescale results in terms of the low-energy scale, i.e. vs T/T_L :

$[T_L = Z V^2/t_*]$

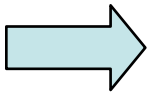
..... same results, now shown vs T/T_L :



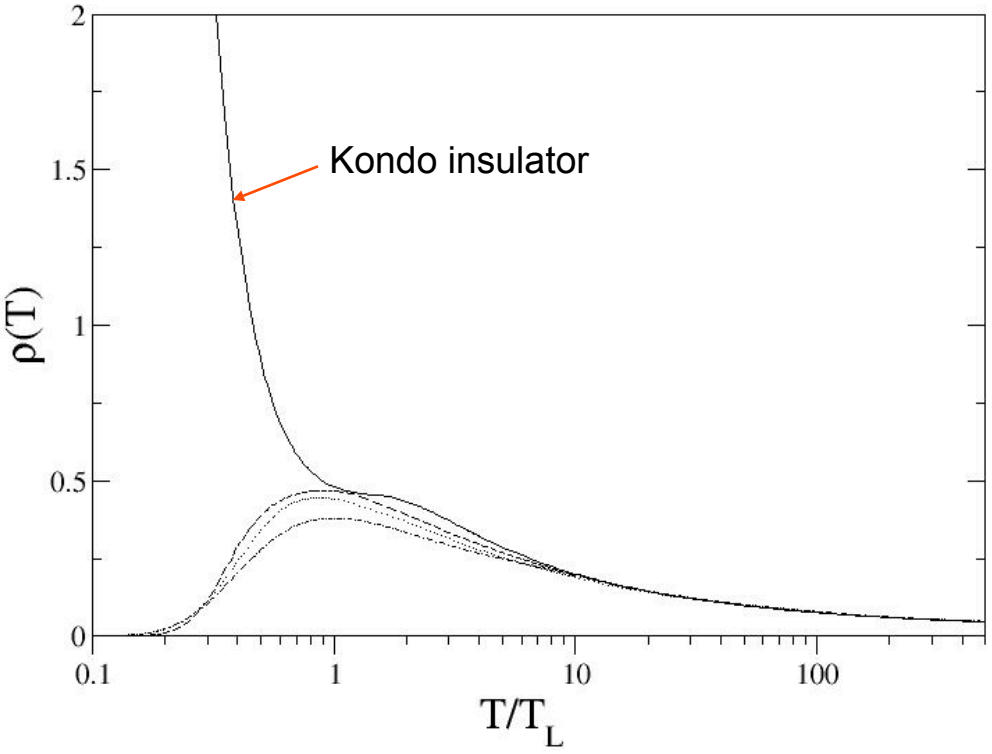
• 'Universal scaling' in terms of the low-energy scale clearly arises, **independent** of the interaction strength U (*and* of the hybridization V).

• Above results for fixed conduction band filling (n_c), as determined by $^2{}_c$:

Obvious question: how does the scaling resistivity depend on the conduction band filling?



$\rho(T)$ vs T/T_L, varying the conduction band filling n_c:

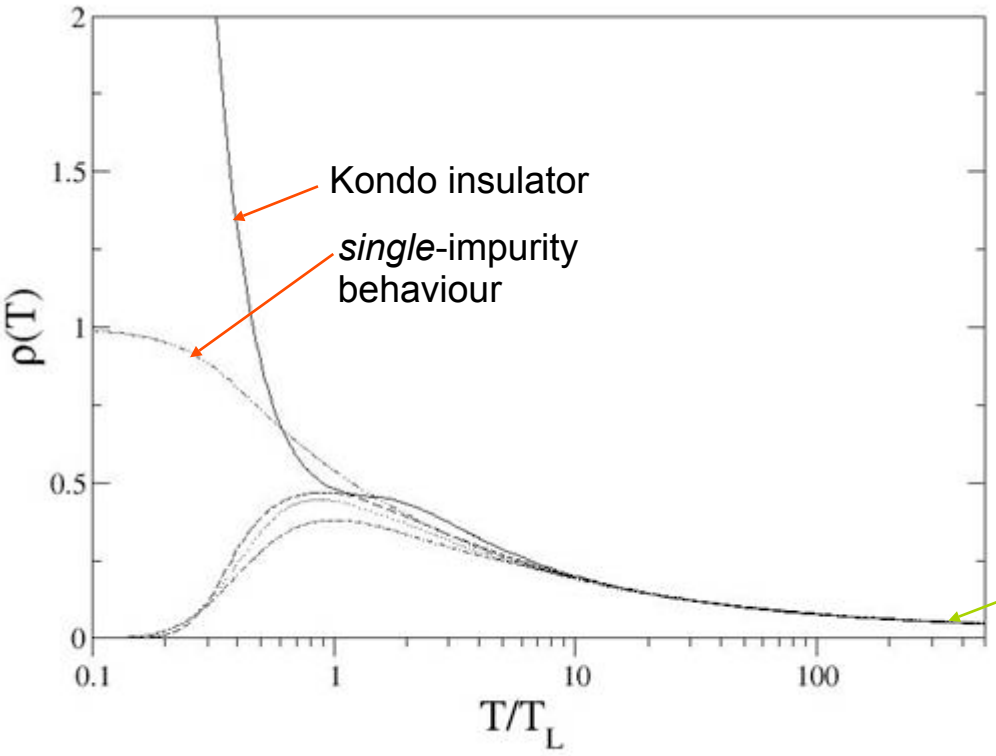


- Same as before: $n_c \simeq 0.7$.
- Now showing: $n_c \simeq 0.7; 0.5; 0.4$:

  'Tails' clearly common to all.

- Including the Kondo insulator --- again, common 'tails'.

$\rho(T)$ vs T/T_L, varying the conduction band filling n_c:



- Origin of the common behaviour --- 'incoherent' *single*-impurity-like scattering (c.f. Anderson *impurity* model [AIM]).

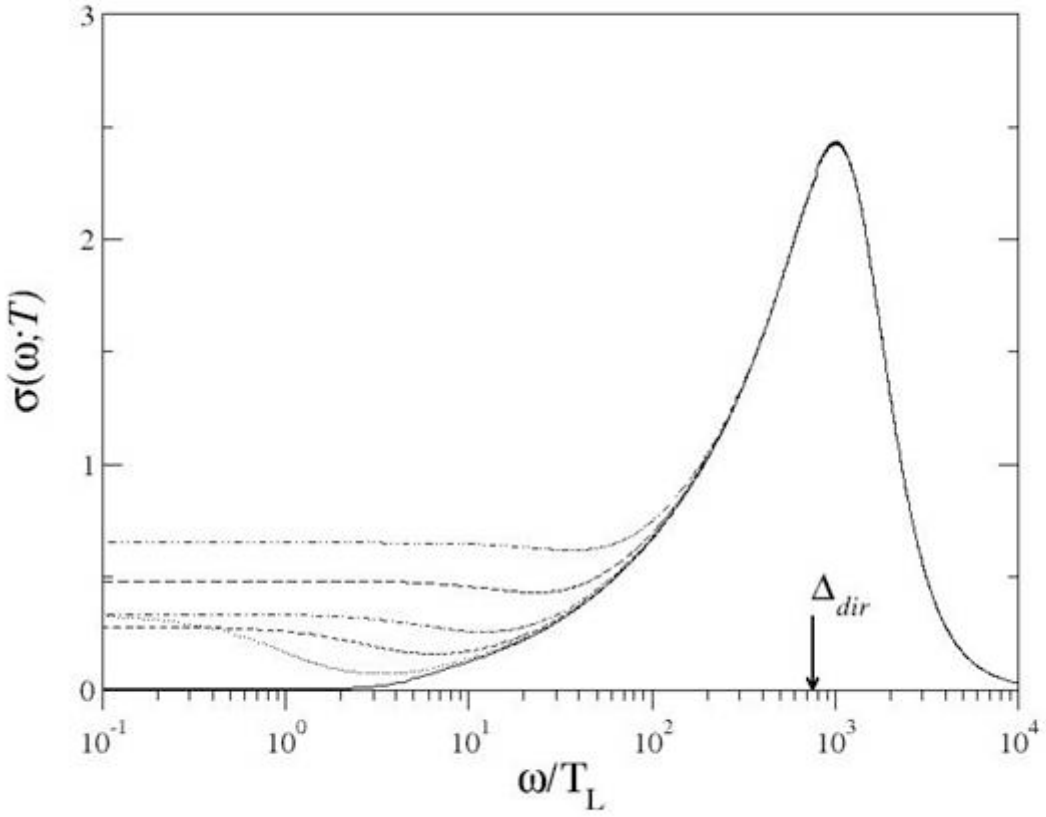
- Note 'slow log tails'; in part'r, for

$$\frac{T}{T_L} \ll 1 : \rho(T) \sim \frac{3\pi^2}{16} \frac{1}{\ln^2\left(\frac{T}{T_L}\right)}$$

--- exact asymptote for S=1/2 AIM.

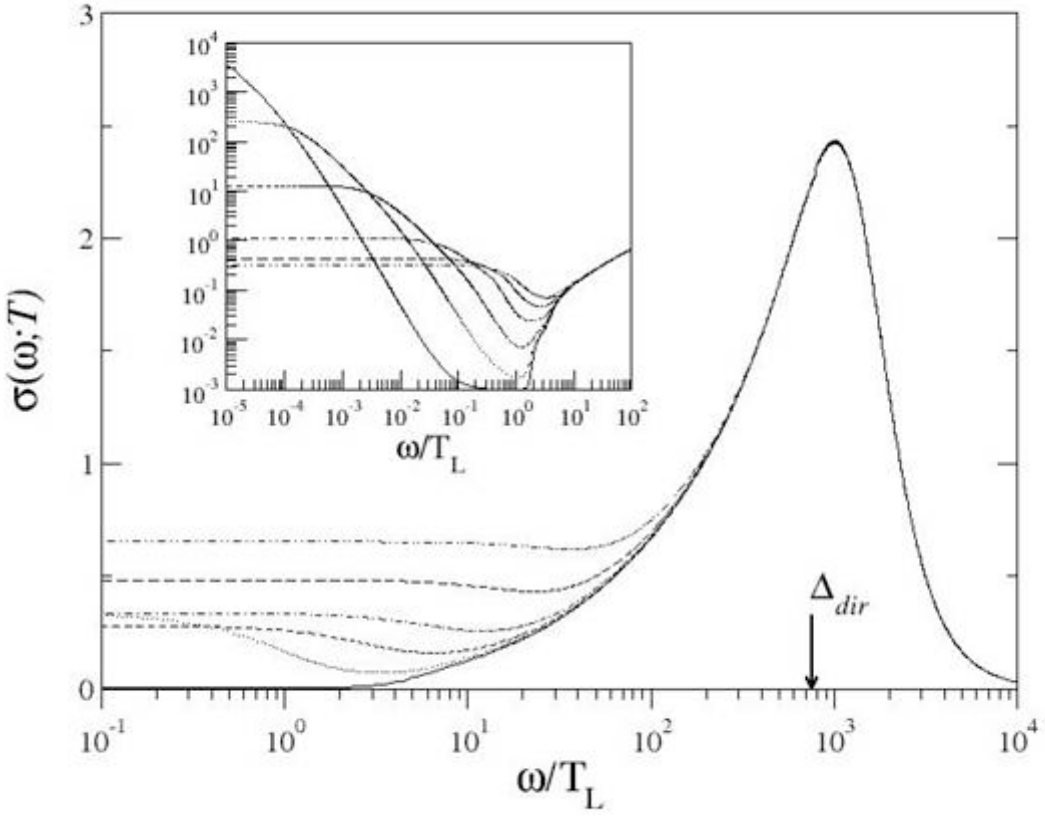
- Note: conclusion arises on comparison of $\rho(T)$ *scaling behaviour* for PAM *vs* AIM. This is *independent* of how the PAM coherence scale, T_L, and the AIM Kondo scale, T_K, depend on 'bare' model/material parameters; it also precludes as such 'two-scale' exhaustion.

Optical conductivity: $\sigma(\omega;T)$ vs ω/T_L



- Strong absorption in vicinity of direct gap Δ_{dir} ($\sim 750T_L$ here).
  --- with continuous absorption at lower energies.
- Strong thermal evolution, shown for:
  $T/T_L = 0; 0.5; 1; 2; 5; 10$
  --- and T_L clearly 'sets scale' for thermal evolution.

Optical conductivity: ¾(! ; T) vs ! =T_L

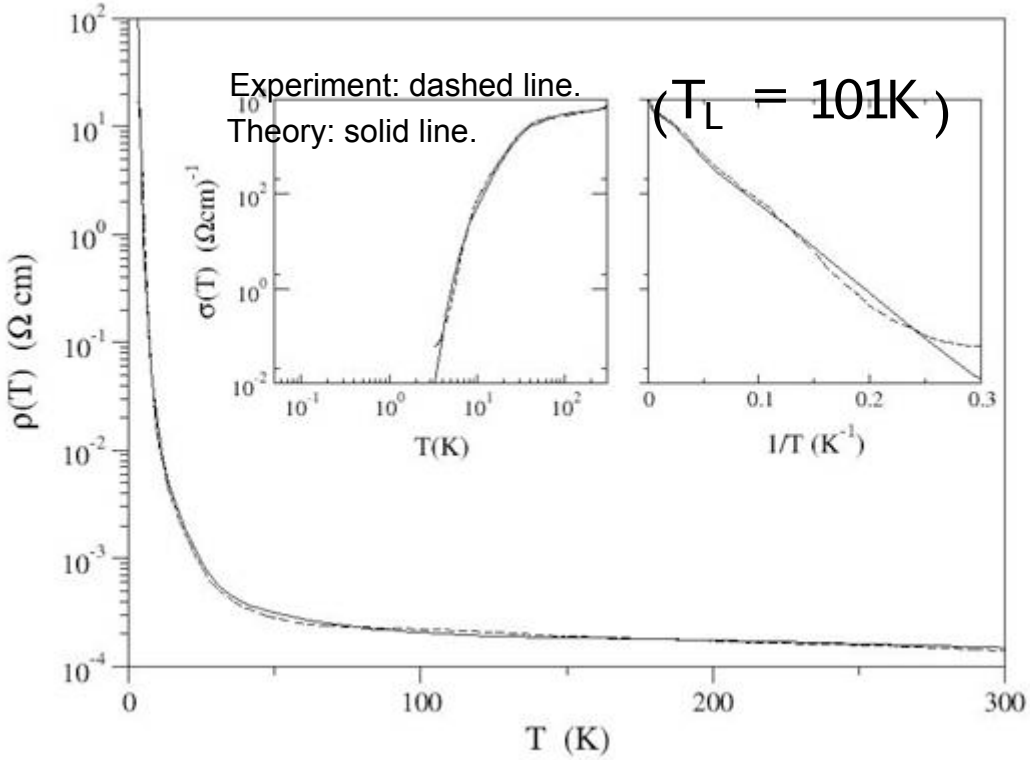


- For lower ! =T_L and T =T_L scales:
  | for T =T_L from 0:02; 0:1 ... 0.5.

  ---- showing Drude absorption,
  its thermal evolution/destruction.

Comparison of theory and experiment.

1. SmB_6 --- a strongly correlated Kondo insulator.

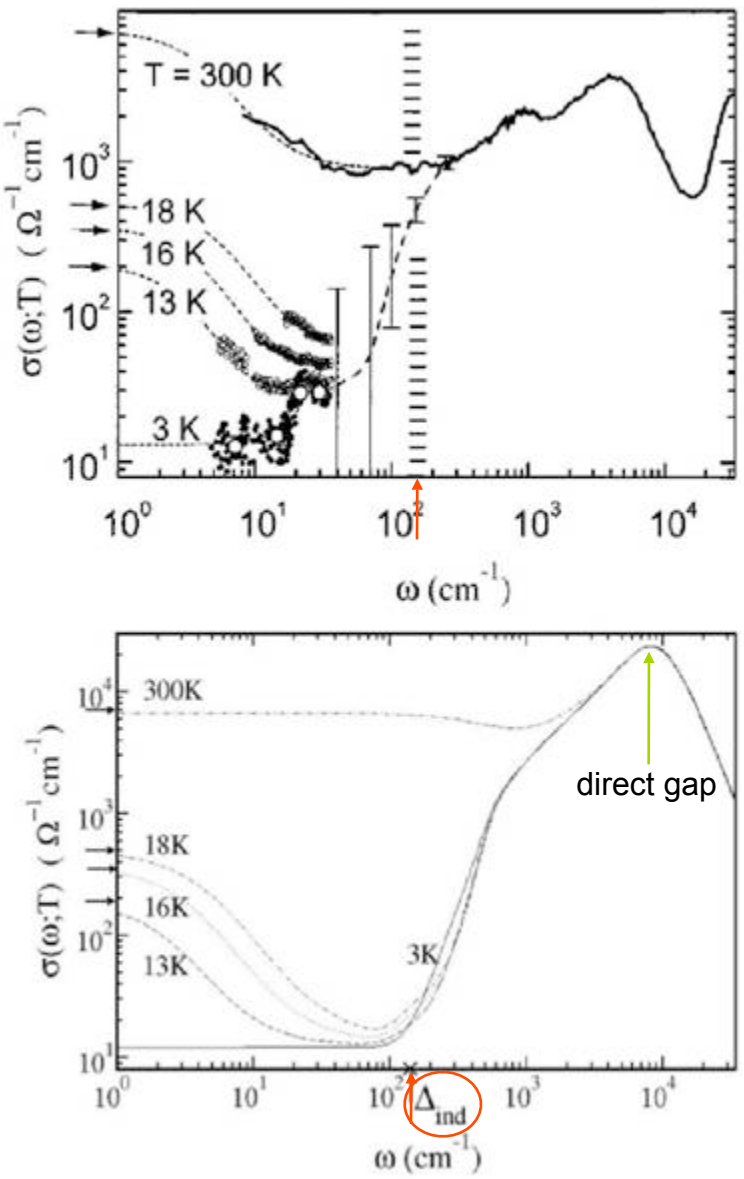


- We compare to expt by exploiting scaling: that ½ ½($\frac{T}{T_L}$): How? …… so predictive. How do theory/expt compare?

- Now move to the optics --- to predict their ! -dependence *and* thermal evolution.

Gorshunov et al Phys Rev B **59,** 18081 (1999).

SmB_6 : **Optics.**



- Level of agreement self-evident.
  Features .....

- One further point: for an insulator, at $T = 0$ there must exist a 'true' gap in the optical conductivity --- the **indirect gap**; given by

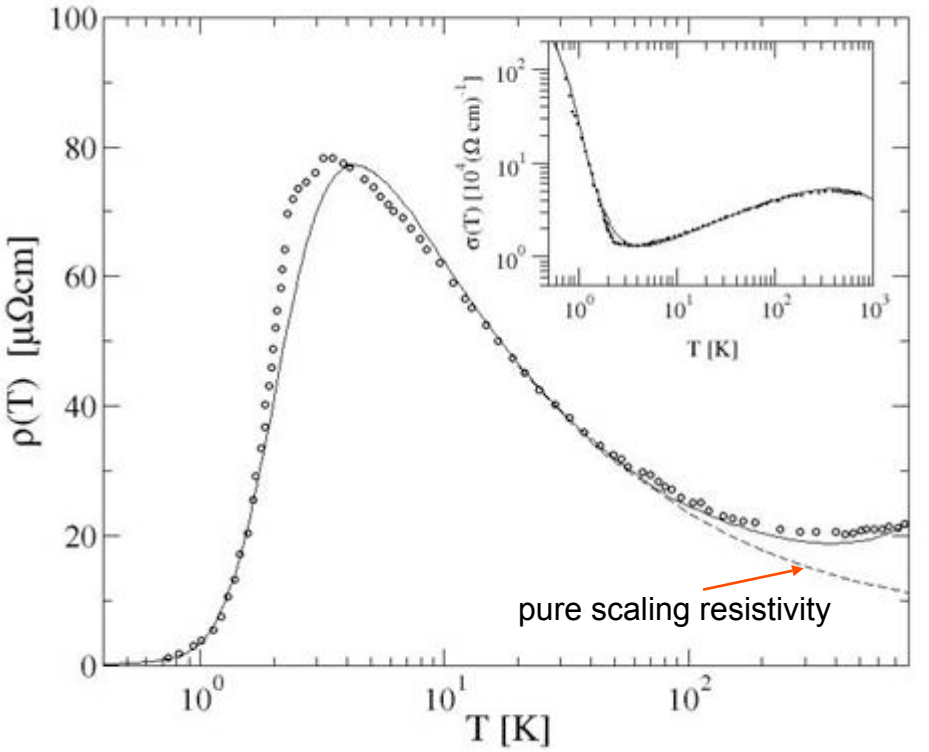
$$¢_{ind} = 2T_L :$$

So since we know T_L ; $¢_{ind}$ follows --- as marked in lower figure, and compared to experiment (upper figure).

Gorshunov et al Phys Rev B **59,** 18081 (1999).

Nanba et al, Physica B, **186-188**, 440 (1993)

2. CeB_6 --- a strongly correlated HF metal.



Sato et al, J. Phys. Soc. Jpn., **54,** 1923 (1985).

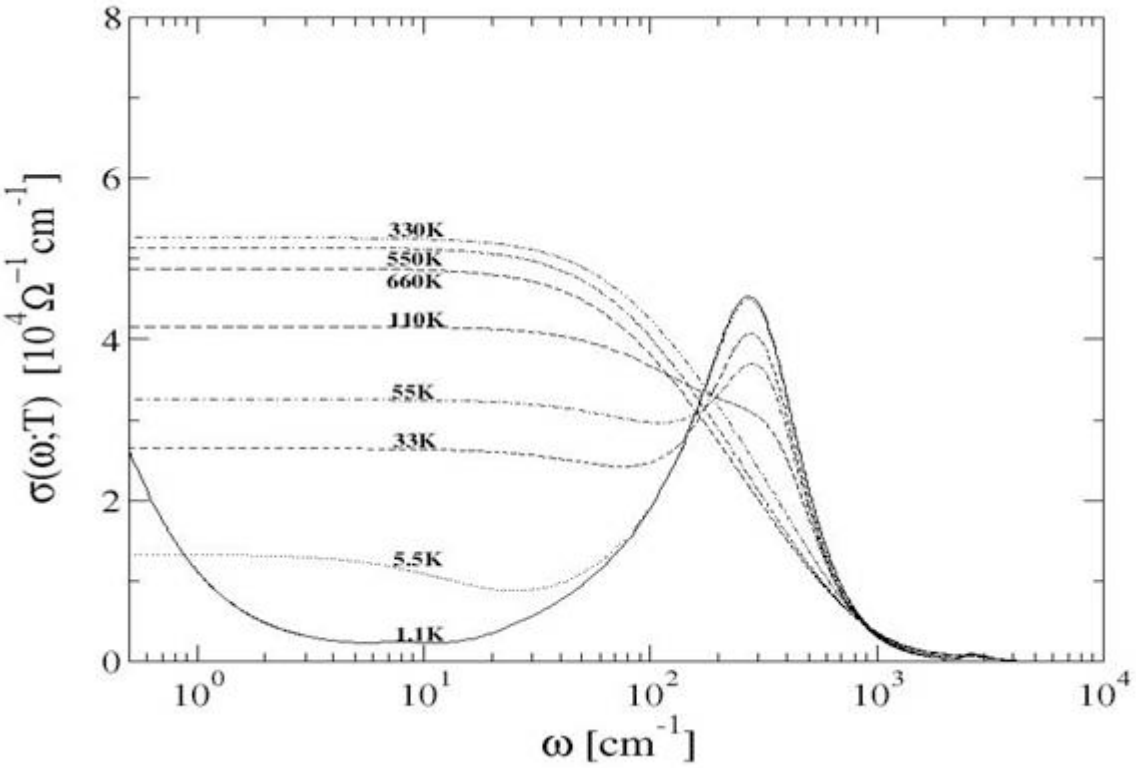
• Resistivity has classic form for a HF metal; its maximum » demarcates the crossover from 'lattice-coherence' to 'incoherent' scattering.

Expt: circles. Theory: solid line.

Can also see the (correct) departure from the pure scaling regime itself, beginning at $T \gg 100K$ ($\sim 20T_L$):

What about optics? Predict .....

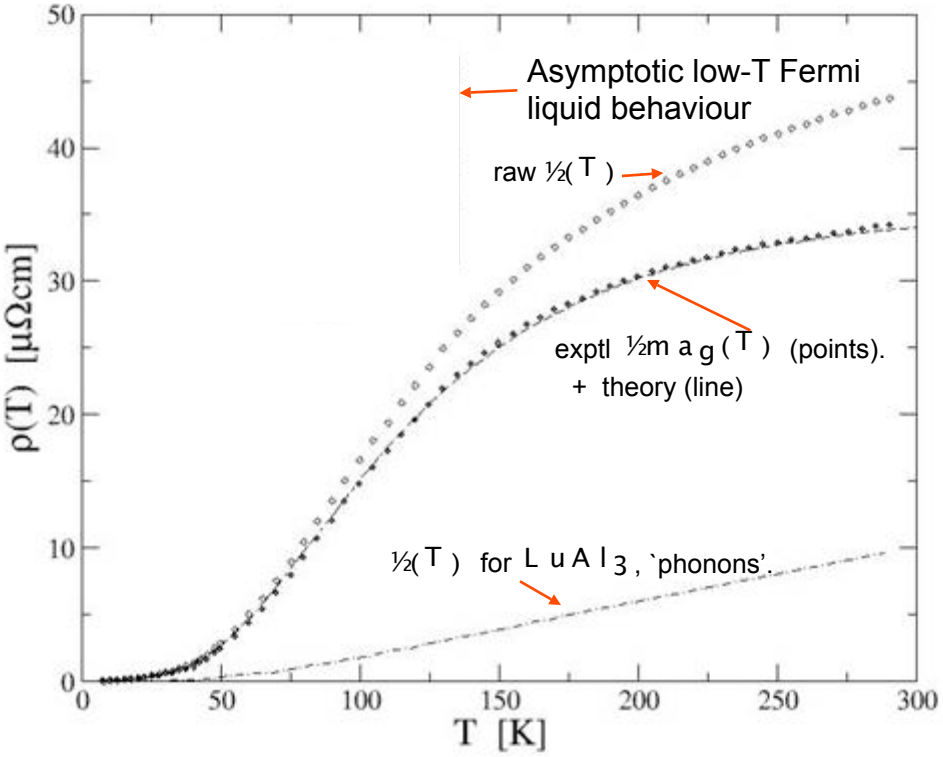
Predicted optics for CeB_6 : --



Optical measurements have been performed ¤; but only at $T = 300K$ and for frequencies ! & 400 cm^{-1} --- are consistent with theory, but to observe non-trivial ! and T-dependences, clearly need measurements at much lower T and down to much lower lower frequencies » 1¡ 10cm^{-1}:

* Kimura et al, Phys Rev B, **50**, 1406 (1994).

3. $YbAl_3$ --- a classic intermediate valence metal.



- Quite distinct from strongly correlated HF metal: $\rho(T)$ increases monotonically with T.

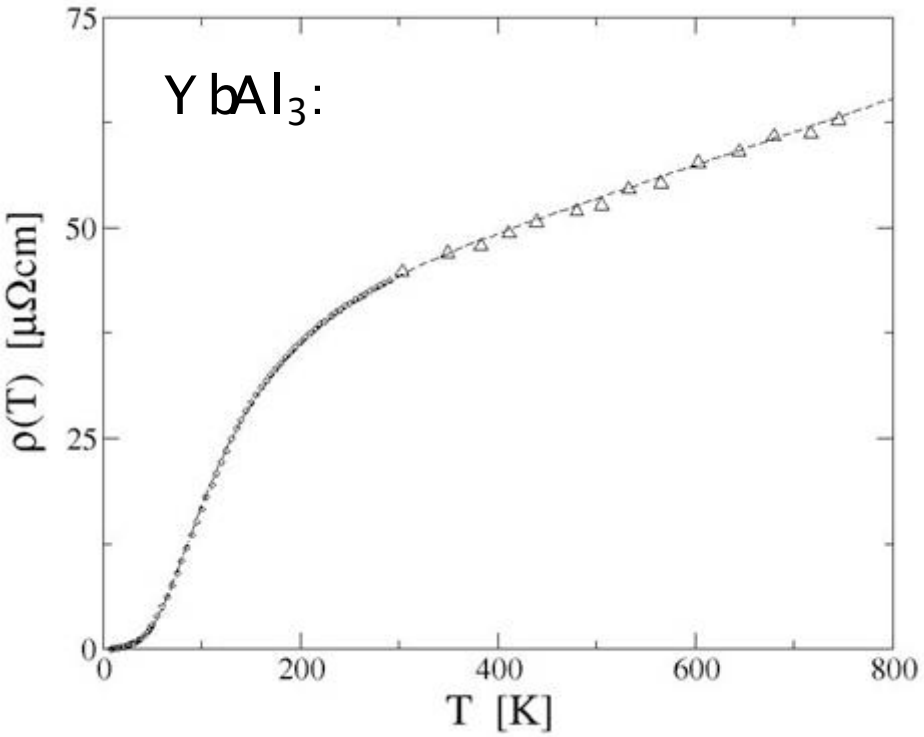
- Characteristic of IV behaviour: f-level ε_f close to Fermi level, so f-levels partially occupied. [Yb valence $Z_v = 2 + n_f \simeq 2.65$]

  But still quite strongly correlated, $T_L \simeq 250$ K :

- Provides good example of Fermi liquid at the lowest T's: ---

  $\rho(T) \propto T^2$; here holding for $T \lesssim 30$K :

Ohara et al, J Alloys Compounds, **323/324**, 632 (2001).



Theoretical resistivity out to T=750 K, compared to experiment.

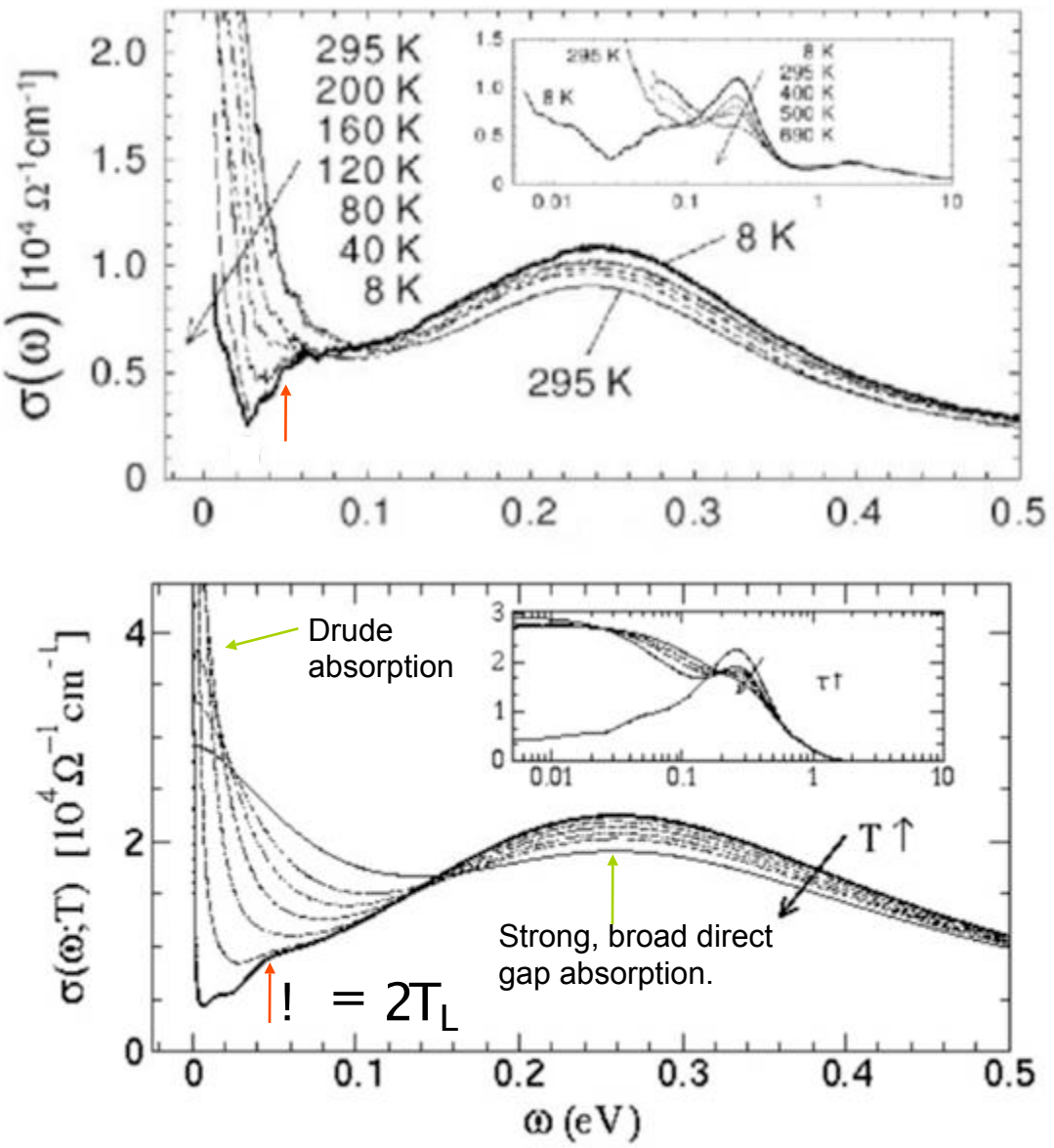
So what of the optics?.....

Rowe et al, J. Phys. D: Appl Phys, **35**, 2183 (2002).

Ohara et al, J Alloys Compounds, **323/324**, 632 (2001).

$YbAl_3$: **Optics.**

Expt: Okamura et al, J. Phys. Soc. Jpn., **73**, 2045 (2004).



- See clearly:

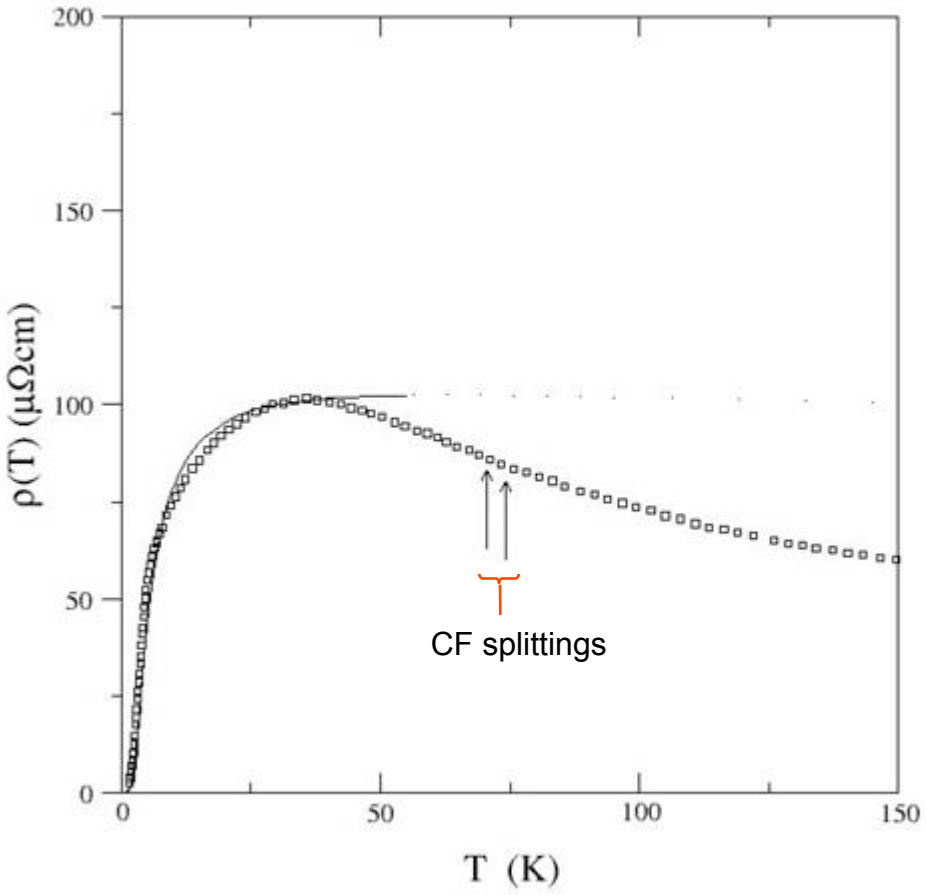
--- low-energy Drude absorption and its strong thermal evolution (+ weak 'pseudogap' at lowest T).

--- Strong, broad direct gap absorption at $! = ¢_{dir}$ ' 0:25eV:

--- Theory predicts a weak 'shoulder' above the pseudogap, at $! \quad ' \quad 2T_L$:'Robust' theoretically.

Seen in experiment, and characteristic of IV behaviour -- e.g. occurs also in the Kondo insulator YbB_{12}:

4. $CeAl_3$ …. a strongly correlated HF metal.



- Much studied system, with 'unusual' optics (below). First, a look at its d.c. resistivity: clearly, theory and expt agree fine up to ca 50K. But not thereafter.

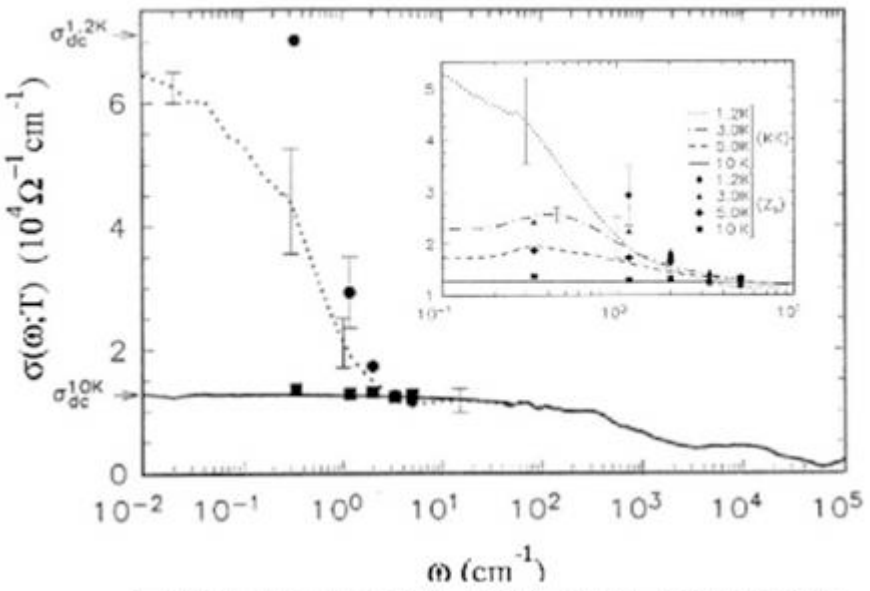
- What's missing in theory
  --- crystal field splitting (by construction). Two such here, at around 6meV = 70K --- pretty low. Behaviour seen thus expected.

Kagayama et al, J. Alloys Compounds, **192**, 239 (1993).

$T_L = 33$ K

$CeAl_3$: **Optics.**

Expt: Awasthi et al, Phys. Rev. B **48**, 10692 (1993)



- See clearly thermal destruction of the characteristic metallic Drude peak (! ¿ 1cmi 1) by T=10K.

- But optics well known to be odd: ------ no strong direct gap absorption, or pseudogap. Can theory account for that?

- Yes. Its origin? ……..

  ..... believe system to have relatively low conduction band filling n_c, but strong hybridization V ; such that n_f ' 1 and strongly correlated: $\frac{m^{¤}}{m}$ ' 700:

  + where theory predicts the direct gap to be is indicated by ↓ --- and indeed no strong absorption there.

Concluding remarks.

• Began with a question: to what extent are real materials captured by the simple/ minimalist PAM? As judged by the transport/optics considered here, answer appears to be pretty well.

• That is so largely because theory handles all experimentally relevant energy/ temperature scales (+ all interaction strengths): if it failed to, little agreement with experiment would arise, even qualitatively.

• Have focussed on 'pure' systems: clearly doping with a non-magnetic ion --- eg $Ce_xLa_{1-x}B_6$ or $Yb_xLu_{1-x}Al_3$ --- then (a) evolve from PAM to AIM physics with decreasing 'x'; and (b) introduce disorder, and hence interplay between disorder and strong interactions. Rich, under study.

Acknowledgements

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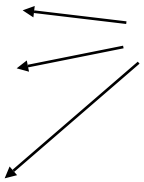

HF and KI work

Martin Galpin, Anne Gilbert, Andrew Mitchell, Matt Glossop.

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