Theoretical description of the high-temperature phase of $YbInCu_4$ and $EuNi_2(Si_{1-x}Ge_x)_2$

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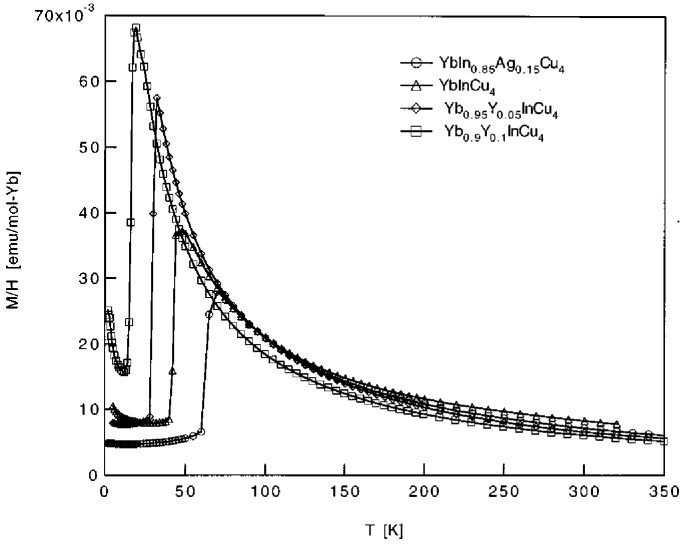
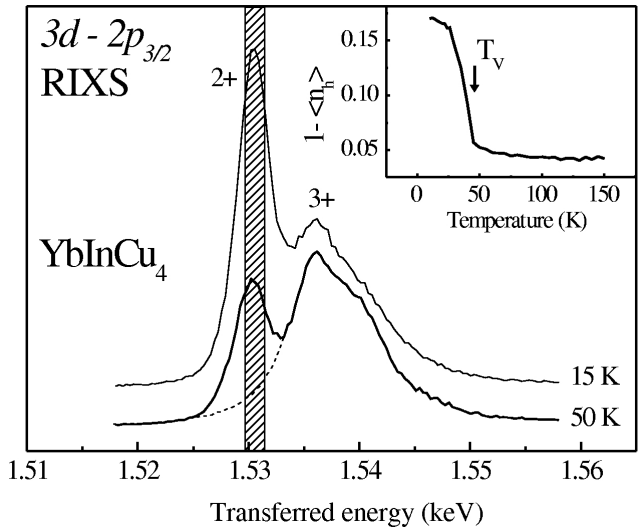
$YbInCu_4$ shows a valence change transition at 42K



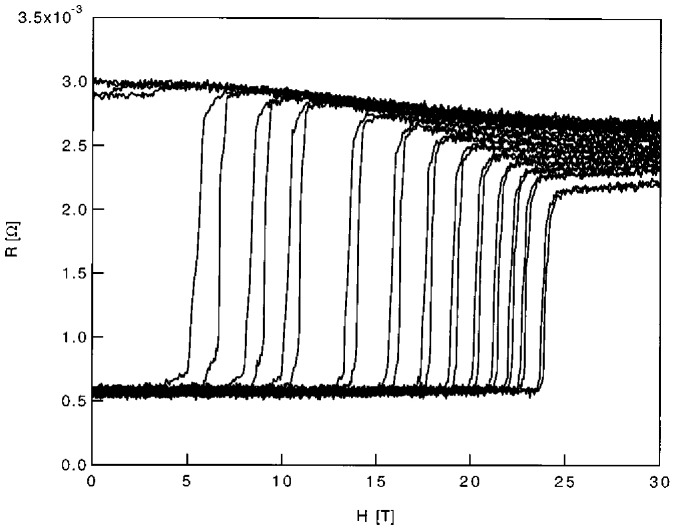


- χ_{spin} shows **Curie-like behavior** with almost the magnitude of the isolated Yb^{3+} moment, but then undergoes a **sharp (first-order) transition** at the valence-change temperature T_V, where the system becomes **paramagnetic**. Doping with Ag increases T_V and broadens the transition; doping with Y reduces T_V and sharpens the transition. *Data from C.D. Immer et al. PRB **56,** 71 (1997).*
- **R**esonant **I**nelastic **X**-**R**ay **S**cattering can be employed to measure the Yb valence as a function of T. In $YbInCu_4$, the **valence undergoes a sharp jump at T_V** as the material enters the low-temperature intermediate-valence state. *Data from C. Dallera et al. PRL **89,** 196403 (2002).*

Effect of a magnetic field

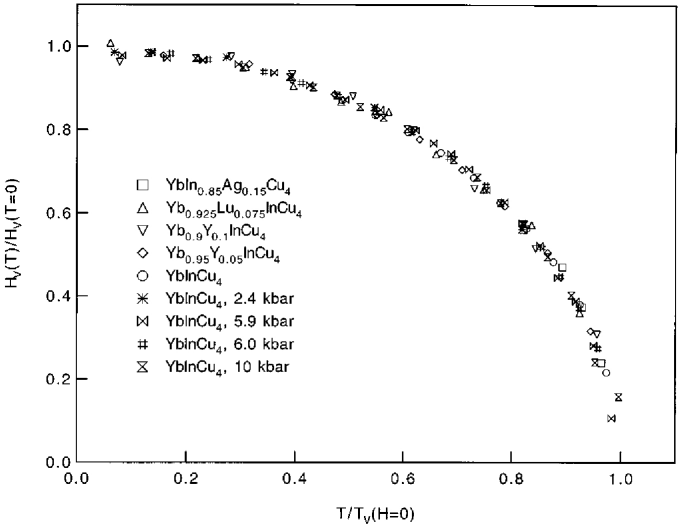
A magnetic field affects the valence-change transition. Increasing H, reduces $T_V(H)$ and increases the high-field resistivity. Hence a **magnetic field favors the high-temperature (local-moment) regime**. The magnetic field also tends to sharpen the transition.

Data from C. D. Immer et al PRB ***56,*** *71 (1997).*

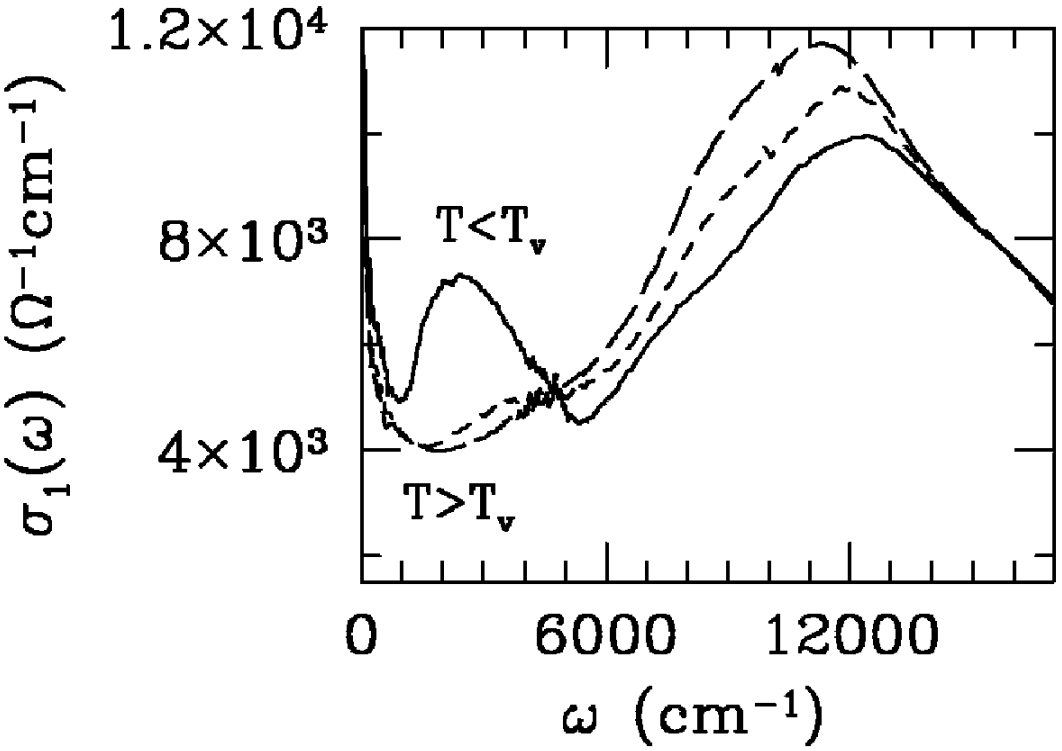


When comparing the magnetic field effects for a variety of materials that are chemically doped with Ag or Y, or are placed under pressure, one finds a **universal relation** between the critical field at T $[H_V(T)]$ and the critical temperature at H $[T_V(H)]$. They have a square-root dependence.

Data from C. D. Immer et al PRB ***56,*** *71 (1997).*



Optical conductivity in $YbInCu_4$

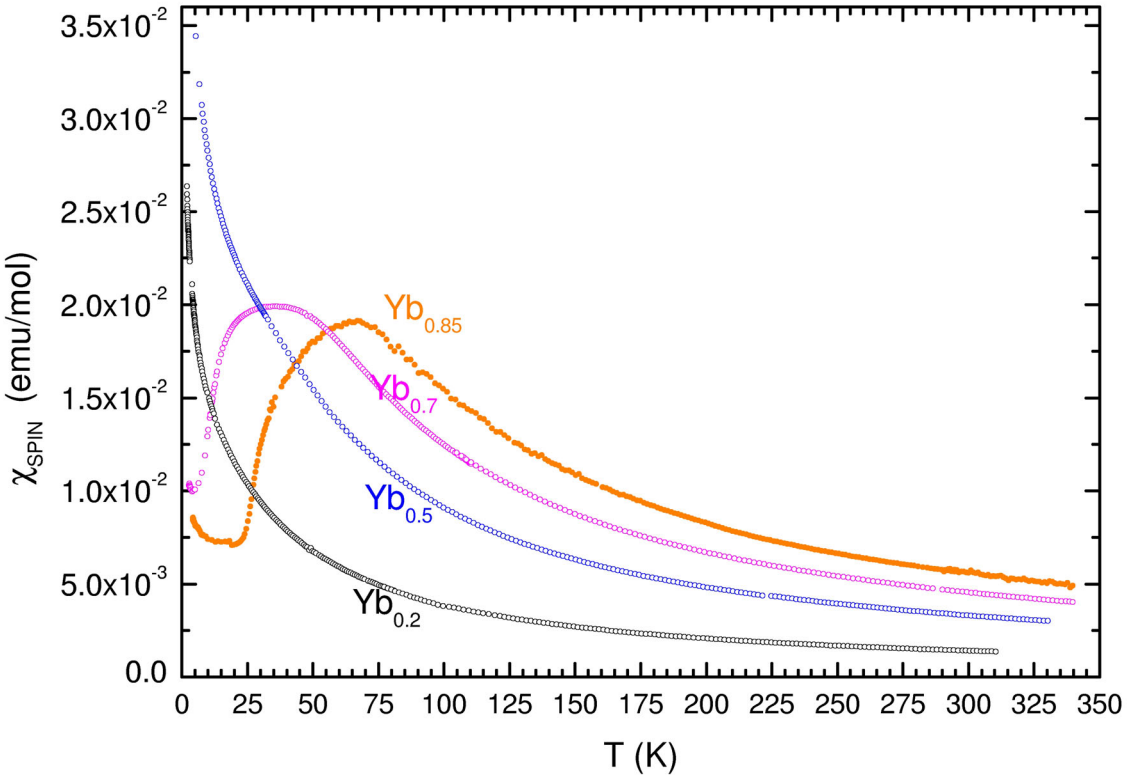


Data from Garner et al, PRB ***62,*** *R4778 (2000).*

- The optical conductivity has interesting behavior as well. As the temperature is reduced towards T_V, the spectral weight is reduced in the charge-transfer peak and slightly enhanced in the mid-IR region. Once T_v is crossed, there is a **dramatic enhancement** of the optical conductivity in the mid-IR region, indicating a **redistribution of spectral weight at the transition**. Hall effect shows an order of magnitude change in carriers at Tv as well.

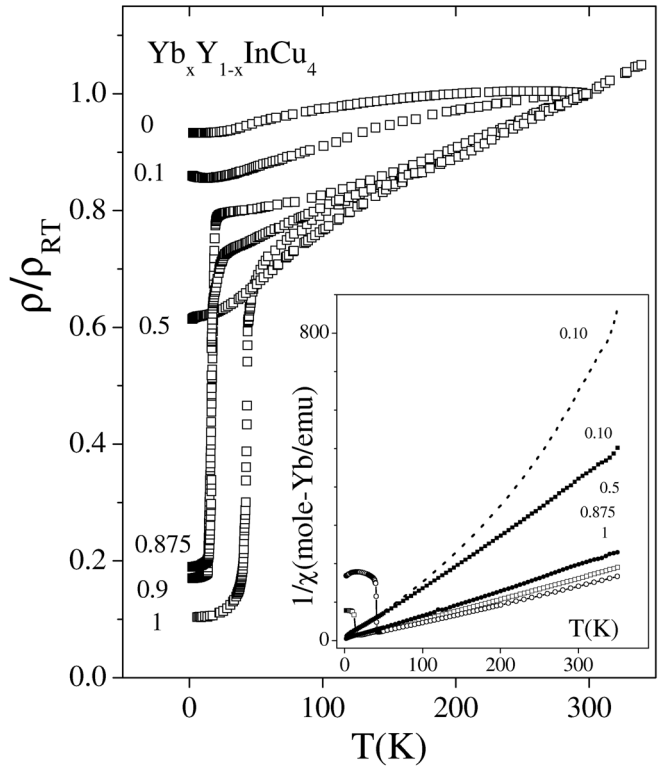
$Yb_xY_{1-x}InCu_4$ has T_V approach 0

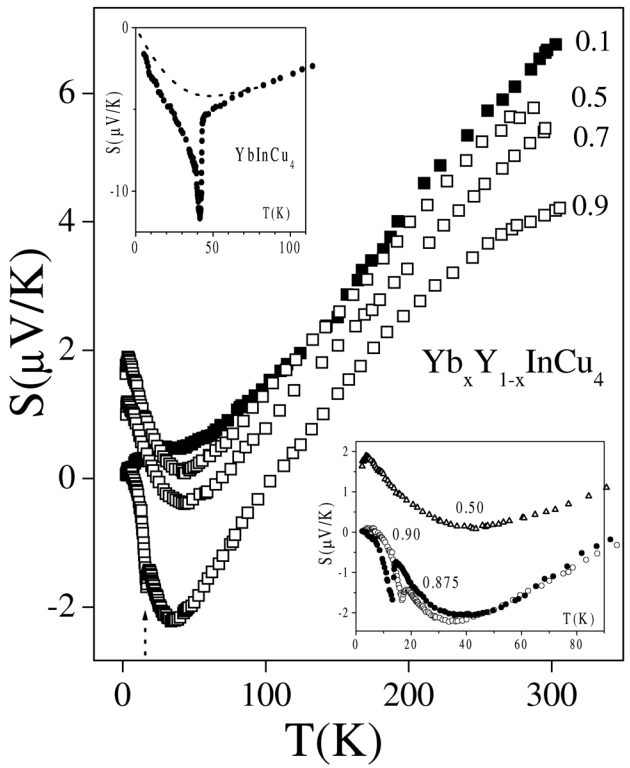
- Note how doping with Y pushes the transition to **smaller** T_V eventually **suppressing the transition** completely.
- This situation allows one to examine the **high-temperature, local-moment regime** without worrying about the physics behind the transition.



Data from I. Aviani, et al (unpublished).

Transport in $Yb_xY_{1-x}InCu_4$





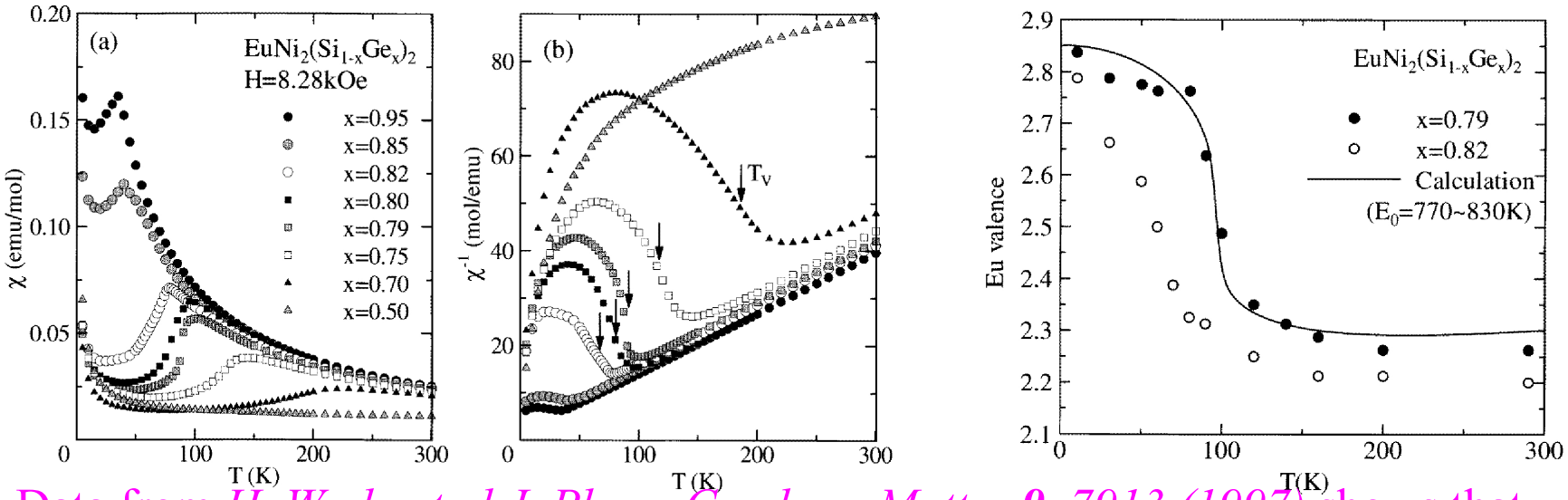
*Data from M. Ocko and J. Sarrao, Physica C **312 & 313**, 341 (2002).*

- The jump in the resistivity is first **reduced then disappears** as the Y doping increases.
- The thermopower has a broad low-energy dip, whose energy scale **is not correlated with T_V** because it does not change with Y concentration.

Summary of Experimental Data $YbInCu_4$

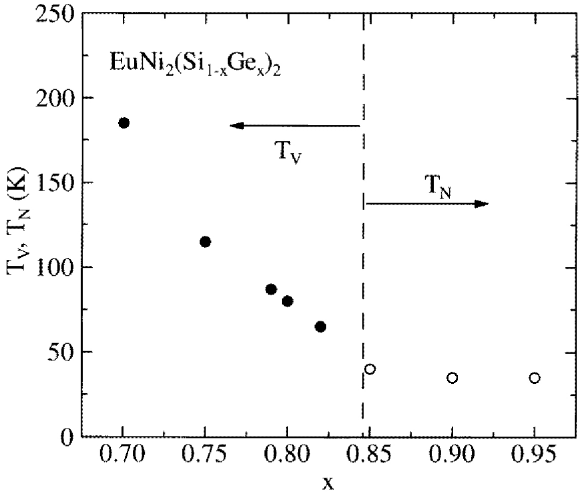
- A **first-order valence-change transition** occurs near 42K characterized by a jump in the Yb valence, a reduction in the resistivity, and a loss of local moments.
- Doping with Ag **increases T_V and broadens** the transition.
- Doping with Y or applying pressure **suppresses T_V** eventually making the transition disappear.
- The electronic density of states are **significantly reorganized** at the transition.
- The transition can also be driven by a magnetic field, which sharpens the transition and suppresses T_V. There is a **universal relation** between the critical temperature and the critical field.

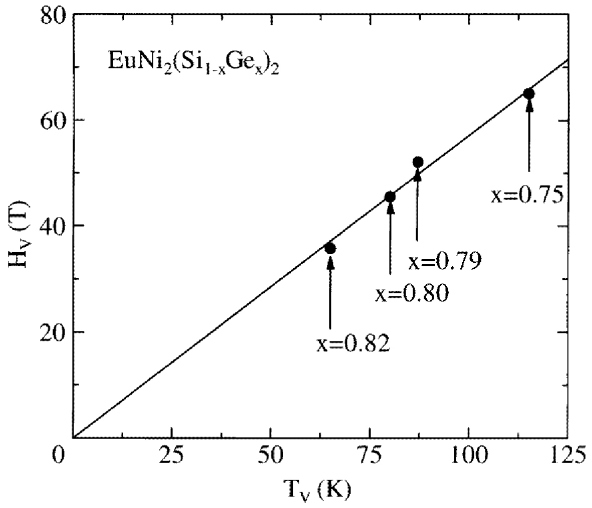
Experimental data on $EuNi_2(Si_{1-x}Ge_x)_2$



- Data from *H. Wada et al J. Phys.: Condens. Matter* ***9,*** *7913 (1997)* shows that there is an **intermediate range of doping x** where the system undergoes a **valence change transition** as well. Outside of this range, the material is **antiferromagnetic** at low T.
- T_V is **typically higher** than in the Yb compounds, and the valence change is **significantly larger** as well (note that recent experiments on M edge rather than L_3 edge show a smaller jump in the valence, but that is believed to be due to surface effects [*Kinoshota et al J. Phys. Soc. Japan* ***71****, 148 (2002)*]).

Phase diagram for $EuNi_2(Si_{1-x}Ge_x)_2$

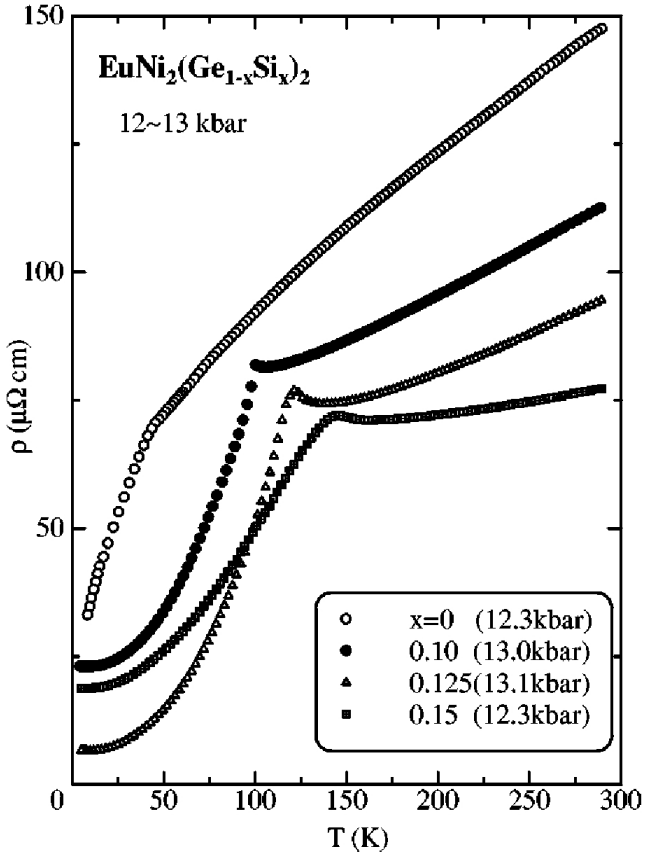




- The phase diagram shows that when the divalent Eu ion is stable, one has **low temperature antiferromagnetism**, but when it is unstable with trivalent Eu, the **valence-change transition occurs**.
- T_V **depends on doping** while T_N is **insensitive to doping**.
- The scaling of H_V with T_V indicates that one **might have a universal relation** between them here as well.
- The energy scales and the valence change appear **to be larger** in the Eu compounds than in the Yb compounds.
- The volume change is **much larger** in Eu compounds.

Data from H. Wada et al., J. Phys.: Condens. Matter ***9,*** *7913 (1997).*

Transport in $EuNi_2(Si_xGe_{1-x})_2$



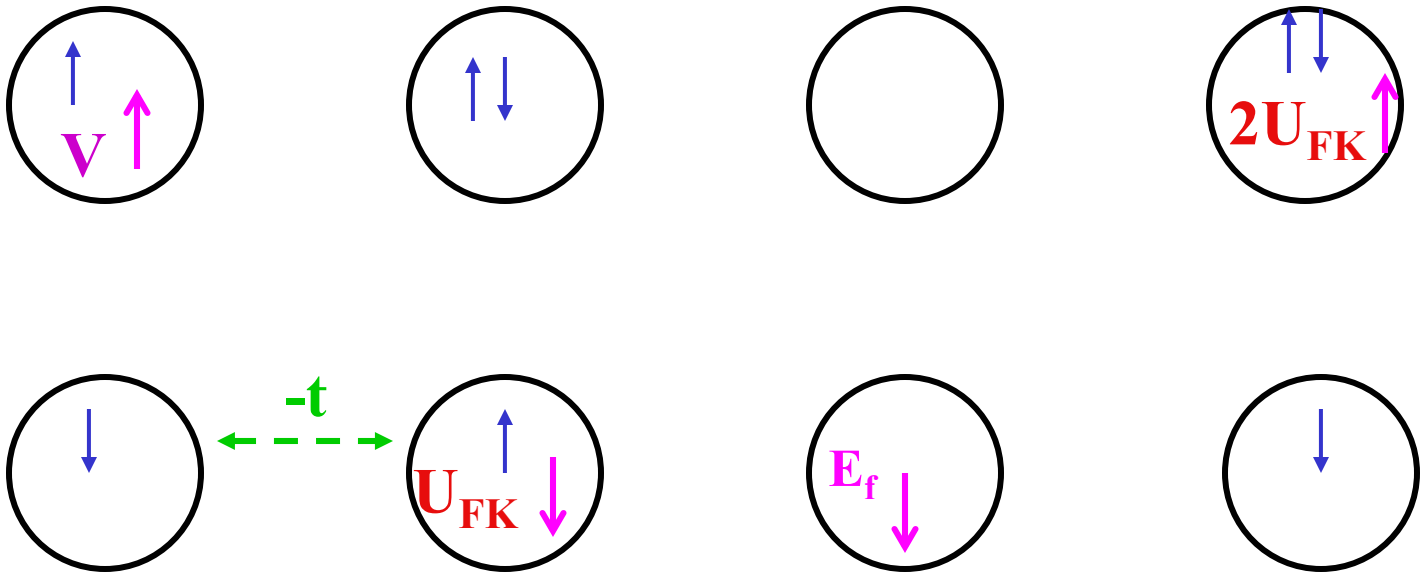
- The transition can also be seen in the **resistivity**, and the signature is enhanced as one increases the pressure.
- The resistivity has a **stronger dependence on T** in the high-temperature phase here than in the Yb compounds.

*Data from H. Wada et al. PRB **59,** 1141 (1999).*

Summary of Experimental Data (Eu compound)

- A valence change transition is seen for **intermediate concentrations** of Ge and Si.
- The temperature scale for T_V and the magnitude of the valence change at the transition are **significantly enhanced** from the Yb case.
- There seems to be the same kind of correlation between transport, and magnetic field effects, as was seen in the Yb compounds. Unfortunately, there have been much fewer experiments performed on the Eu compounds than on the Yb compounds (transport is particularly difficult due to the volume change).
- The correlation of H_V with T_V indicates that a **universal scaling relation** may hold here as well.

The Periodic Anderson/Falicov-Kimball Model



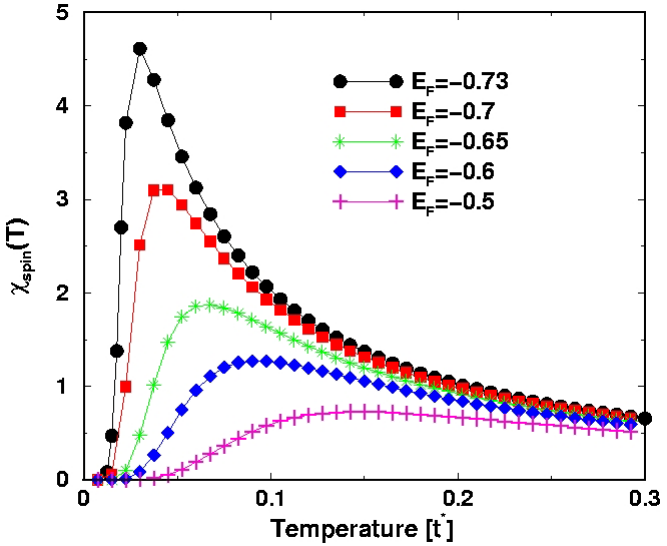
•Conduction electrons **hop** (-t) between nearest neighbors. Localized electrons **cannot hop**, but they **hybridize** with the conduction electrons on the same site (V). The localized electrons have a **site energy** E_f relative to the conduction band center. The Coulomb interaction between two localized electrons is infinitely large, so **no more than one** localized electron can sit on a lattice site at any time. The **Coulomb interaction** between localized and conduction electrons, U_{FK}, is the only Coulomb interaction taken into account.

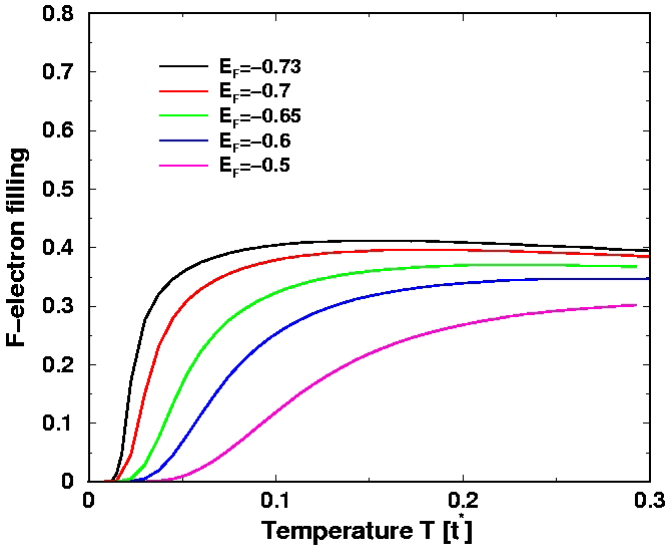
High-Temperature phase

- We focus on the lesser studied **high-temperature phase**. Here we have **local moment formation**, and **poor metallic behavior**. There are **no signs** of the usual Kondo anomalies. Hence it is a good approximation to **neglect the hybridization V** and describe the **pure Falicov-Kimball model**.
- The FK model can be solved **exactly** in the infinite-dimensional limit using dynamical mean field theory. Imaginary axis and real axis data are straightforward to calculate.
- Even though V is ignored, the FK model **does have a valence change transition as a function of T** for some values of the parameters.
- ***This allows one to study the valence-change materials with a simple many-body problem!***

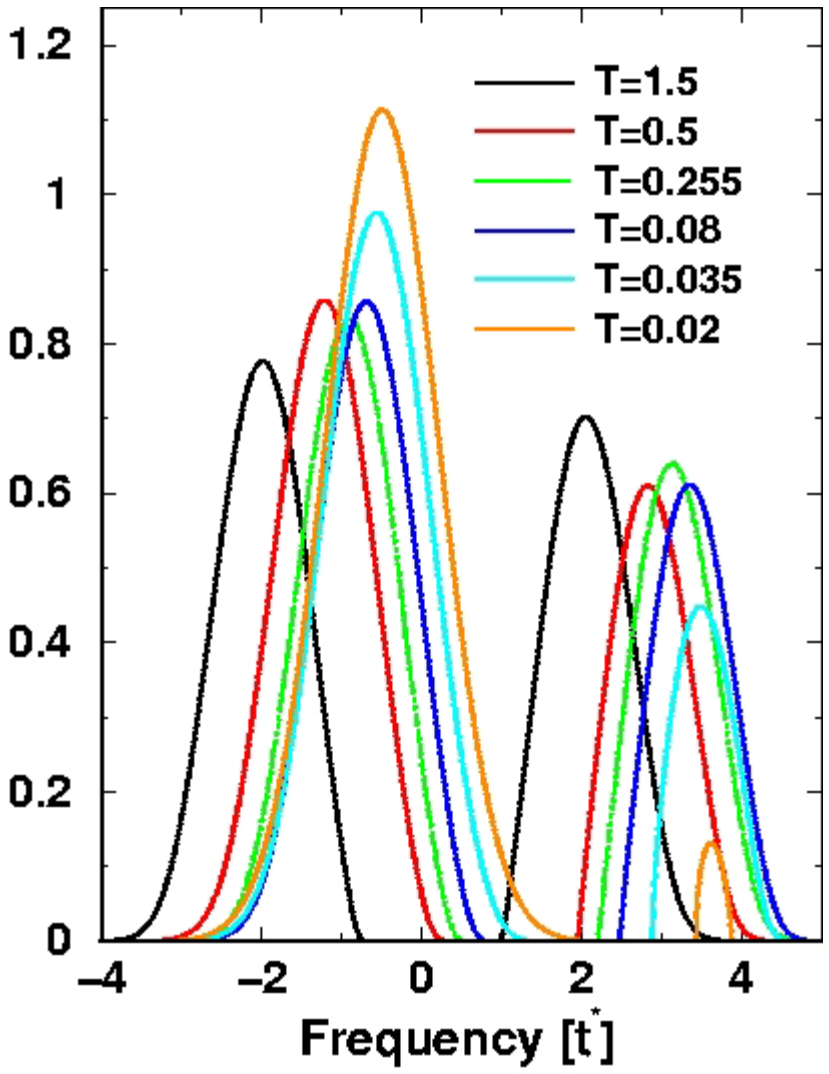
Magnetic response and T_V

- If we choose E_f to lie above the chemical potential as T approaches zero, then there will be **no localized electrons in the ground state**. But if E_f is close to the chemical potential, then the localized electron levels will be **entropically favored and have a significant occupation as T is increased**. Each local moment has a **Curie-like magnetic response** in the FK model, because the localized electrons interact with the conduction electrons only through a charge-coupling; *i.e.* there are no Kondo-like effects.
- Note how the FK model captures the shape of χ_{spin} due to the variation of $n_f(T)$.





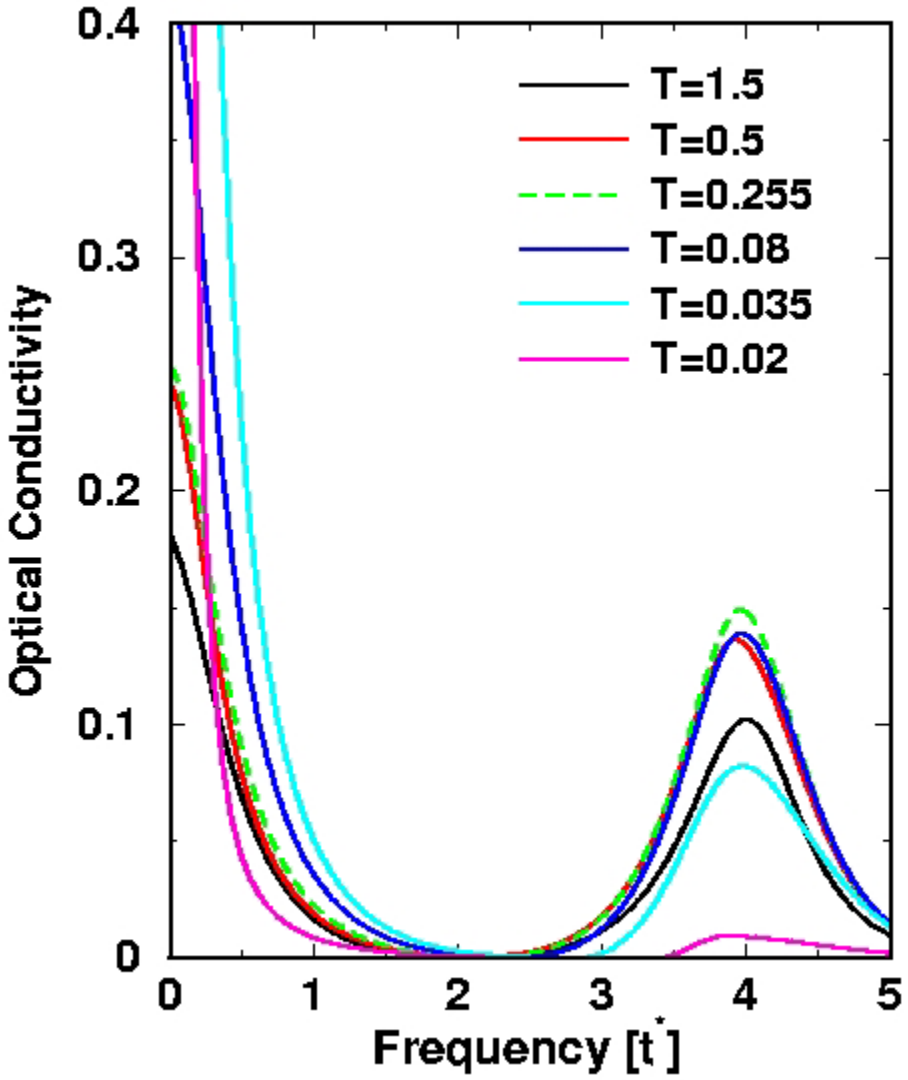
Density of states develops a gap



- We believe that one of the important physical features of the high-temperature phase is that the interacting conduction electron **DOS develops a gap** at the chemical potential which determines high-T transport.
- The gap occurs due to the **interaction of the conduction and localized electrons**, and it moves to the chemical potential as the localized electron concentration increases.
- This is **consistent with Hall data** which measures the number of carriers to be **0.07 in the high-T phase** and **2.2 for low-T**. Theory is not yet sophisticated enough for a direct comparison with photoemission (which also sees DOS changes).

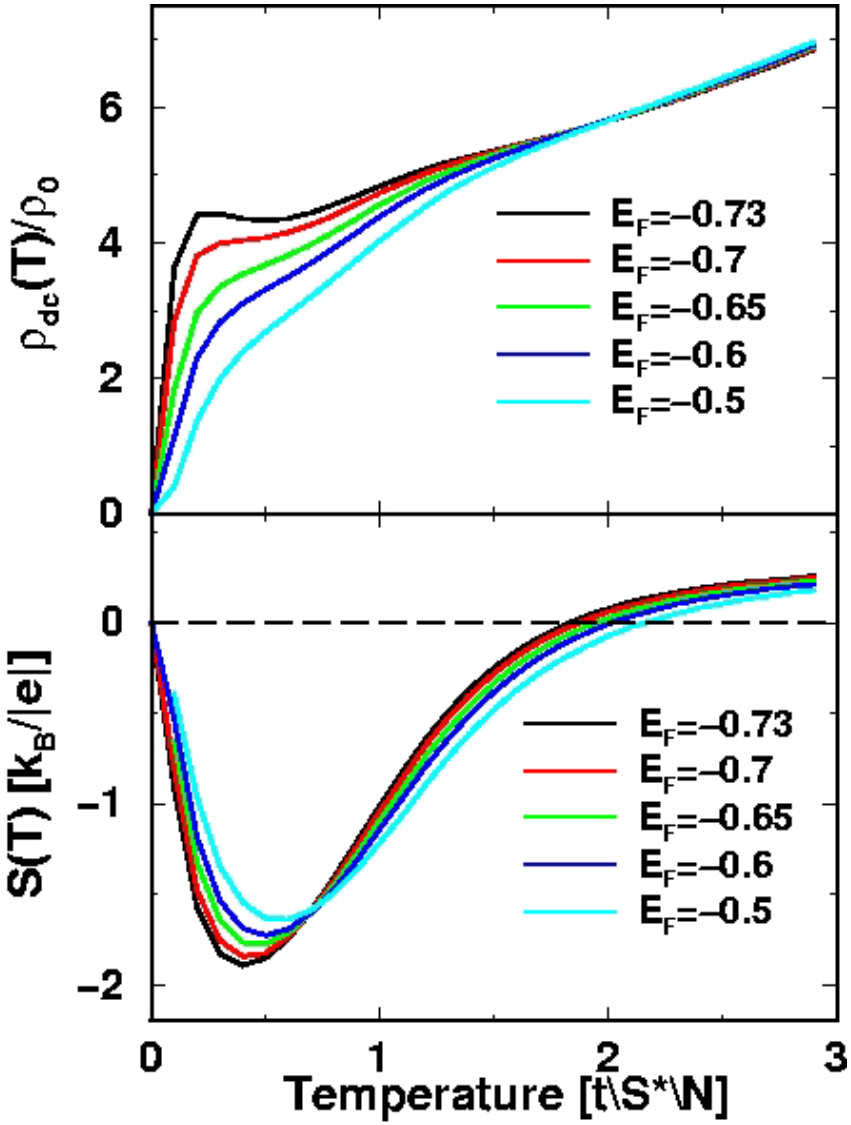
Optical conductivity reflects DOS changes

- The optical conductivity shows a **low-energy feature** and a **charge-transfer peak** at high temperature due to the gap in the DOS.
- As T is lowered, spectral weight **transfers** from the charge transfer peak to a Drude-like feature, but the charge-transfer peak **disappears as T approaches 0** due to the lack of localized electrons in the FK model.
- If the localized electrons, that are present in the experimental low-temperature phase, can couple to light via the renormalized hybridization, then one might **expect an additional mid-IR peak** as seen in experiment.



J. K. Freericks, Georgetown University, Intermediate Valence talk, 2002

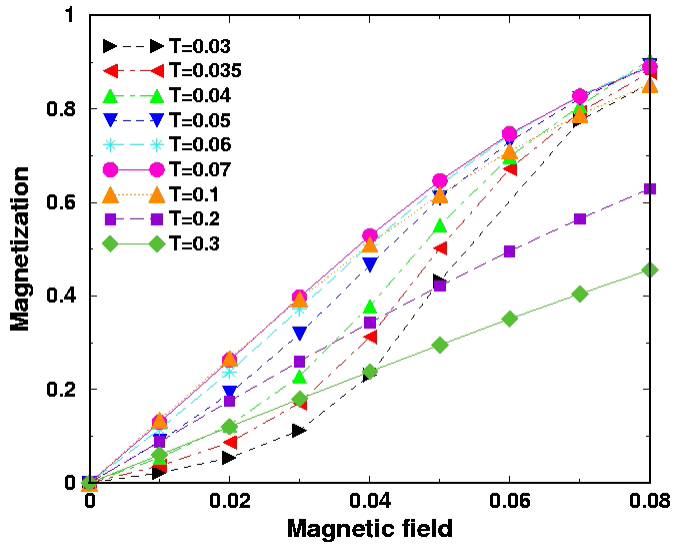
DC transport shows semimetallic behavior

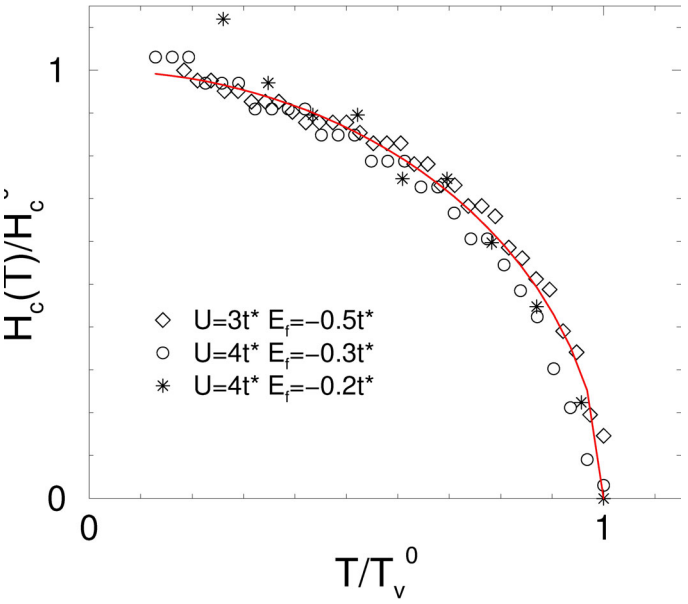


- The resistivity has a **sharp increase at T_V** which **remains flat for a wide range of temperature** in the high-temperature region.
- The thermopower has a **large dip at a temperature T^*,** that is typically larger than T_V, and is **determined by the gap in the DOS**.
- ***Hence the high-temperature transport anomalies are determined by a gap in the single-particle DOS, and not by Kondo-like scattering.***

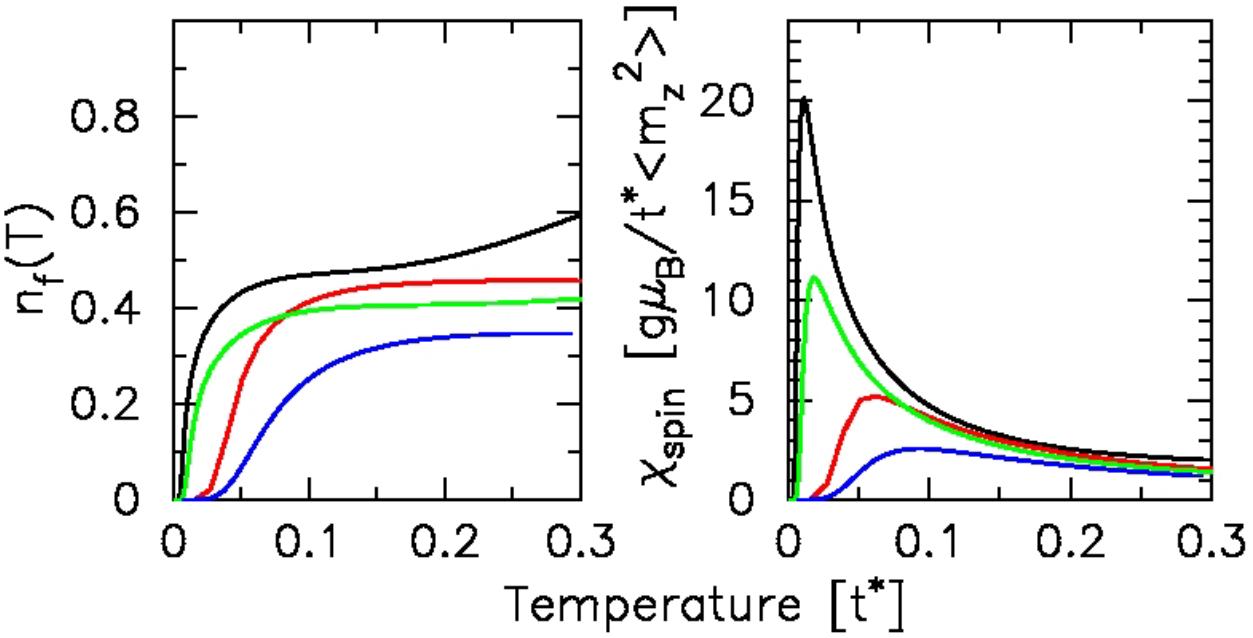
Effects of a magnetic field

- The magnetization displays a **metamagnetic response** in a magnetic field H, when one is below $T_V(H=0)$.
- The metamagnetic transition determines $H_V(T)$.
- A plot of $H_V(T)/H_V(T=0)$ versus $T_V(H)/T_V(H=0)$ shows **the same square-root-like behavior as seen in experiment**.
- ***Hence the FK model appears to capture some of the energetics of the ground state!***



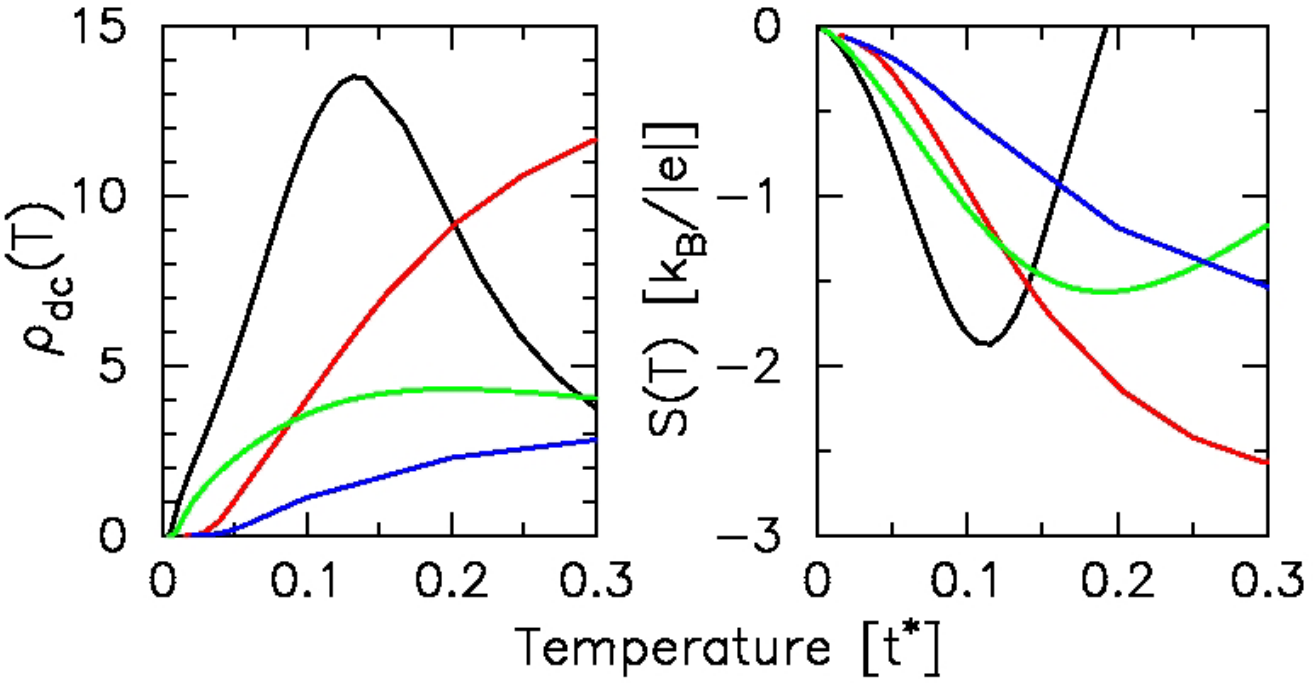


Localized electron degeneracy



- Since the localized electron levels are usually degenerate (eightfold for $YbInCu_4$), and since the occupation is entropically driven, one would expect a **sharper transition** for the higher degeneracy case.
- Indeed, this is seen when we compare solutions with N=2 and N=8. The high-temperature value for n_f is larger and the **crossover is sharper** as the degeneracy increases.
- Similar effects are seen with the magnetic susceptibility as well including **a decrease of T_V**.

Effect of degeneracy on transport



- The resistivity has a **much larger peak** in it as a function of T when the degeneracy is increased. This is **not observed** experimentally.
- The thermopower **sharpens** somewhat, but the magnitude of the peak is about the **same** when degeneracy increases.
- Perhaps the resistivity will be handled properly when **crystal-field and spin-orbit effects are included**.

Summary of theory (successes)

- Theoretical results are **qualitatively similar** to experimental results measured in the high-T phase.
- It appears that the transport anomalies are **most likely due to the development of a gap** in the single-particle DOS as a function of T due to the large FK interaction.
- Universal scaling of the metamagnetic and valence-change transitions **indicate that the theory can capture some of the properties of the ground state and the valence change transition**.
- Detailed comparison with experiment for the different compounds will require a **generalization of these results to include crystal-field and spin-orbit-coupling effects**, which can be handled in a straightforward way with these techniques.

Summary of theory (failures)

- The predicted valence change of the localized electrons is **too large** in the FK model.
- The FK model **does not properly describe** the low-temperature phase (in particular there is **never any Kondo effect or quantum-mechanical intermediate valence**).
- Localized electron dynamics **are not included** in transport properties.
- Adding electron degeneracy reduces the agreement with experiment for some properties.
- A complete model for these materials requires solution of the **full periodic Anderson/Falicov-Kimball model**. In the high-temperature phase the **hybridization renormalizes to zero**, and in the low-temperature phase the **FK interaction renormalizes to zero.**

Conclusions

- Showed how one can describe the **high-temperature anomalies** and the **energetics** of the valence-change transition with the Falicov-Kimball model. The anomalous magnetic properties are determined by the **loss of localized electrons at low T**, and the transport anomalies are associated with **a gap formation** in the noninteracting DOS.
- Our theoretical results produce most of the experimental results found in **$YbInCu_4$** and **$EuNi_2(Si_{1-x}Ge_x)_2$** at high temperatures.
- More work needs to be done to properly describe the **low-temperature phase** and provide a **quantitative model** for these materials.