# Theory of $\delta$ plutonium

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# Physical Picture of $\delta$ Plutonium

LDA puts the f electron bands at the Fermi level which as a result can gain hybridization, to take the advantage of this hybridization LDA produces an equilibrium volume

$$\frac{V_{lda}}{V_{eq}} = .7$$

Extreme case of ”LDA contraction”

U [Rn] $5f^3 6d^1 7s^2$

Pu [Rn] $5f^3 6d^1 7s^2$ but 6d state is very close

In the solid we have $Pu^{+++}$ i.e. an $5f^5$ configuration with the rest of the electrons in a broad spd band

Correlations are strong and we need to include the Hubbard U. V alues in the literature U=4, 5 ev. U reduces the energy gain from the hybridization (blocking effect of Coulomb repulsion) . Therefore the volume will increase relative to the U=0 situation

All the f electrons are equivalent the f electrons are neither localized nor itinerant, they fluctuate dynamically between different configurations

A large fraction of the spectral weight of the f electron is well below the Fermi level (localized part of the electron) while some fraction of the spectral weight resides near the Fermi level (itinerant component)

Connection with earlier work and alternative pictures

B. Johansson, Philos Mag, 30, 469 (1974)

Dynamical Mean field picture of the order paramter landscape

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# Ground state predictions for $\delta$–Pu using LDA+U (S. Savrasov)

Employ state-of–art electronic structure algorithms to eliminate unnecessary discrepancies:

Highly accurate full potential linear–muffin–tin orbital method

Allow spin–orbit coupling together with spin–polarization

Treat all low lying semicore states together with valence states

Use most recent generalized gradient approximation (GGA) for exchange and correlation.

# Physical Picture of $\delta$ Plutonium

The f electrons form a multiplet of the $5f^5$ configuration with L=5, S=5/2

However the spin orbit coupling is very strong and substantially reduces the total moment.

Simplistic view $M_S = 2\ \mu_B\ 5/2 = 5\ \mu_B$

$M_L = -3 - 2 - 1 + 0 + 1 = 5\mu_B$

*A nearly complete compensation of spin and orbital moments occurs* in $\delta$–Pu.

$\delta$–Pu. is strongly correlated but the effects of correlations are not manifest in the q=0 magnetic susceptibility because of strong cancellations betweenspin and orbital moment

There is no magnetic long–range order so the correlations are dynamic in nature

Ideal system to implement realistic DMFT

Evaluation of the moment should take into account a) Spin J=5/2 atomic configuration, b) residual crystal field splitting into $\Gamma_7$ and $\Gamma_8$ c) hybridization, i.e. Kondo screening by s-p-d electrons

The residual degeneracy of these configurations accounts for the large resistivity and the specific heat.However, the

heavy fermion behavior should not appear in the magnetic susceptibility.

*Fat bands* (Jepsen, Andersen, 1997)

Visualizing partial characters of the one–electron states

$$\psi_{kj}(r) = \sum_{L\tau} A^{kj}_{L\tau} \phi_{L\tau}(r, \epsilon_{kj})$$

$\int \cdots \int$ where the coefficients

$$\left| A^{kj}_{L\tau} \right|^2$$

carry required information about partial content of every state $kj$ with the normalization

$$\sum_{L\tau} \left| A^{kj}_{L\tau} \right|^2 = 1$$

Fattening the band $\epsilon_{kj}$ proportionally to $\left| A^{kj}_{L\tau} \right|^2$ brings a required visualisation

# Work in Progress

Calculation of one electron Spectra

Transport Properties

# Future Work

Effects of Impurities

Extensions of DMFT to evaluate U internally

Phonons Linear Response

Calculations away from saddle points, reaction rates?