Orbital order and Hund's rule frustration in Kondo lattices

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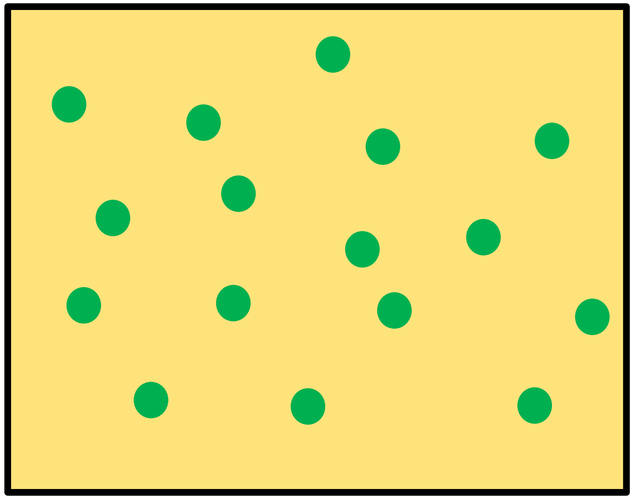
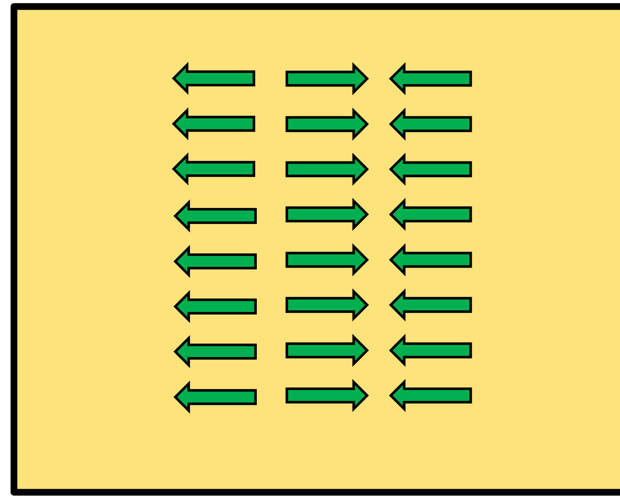
advancing the frontiers

Competing behavior

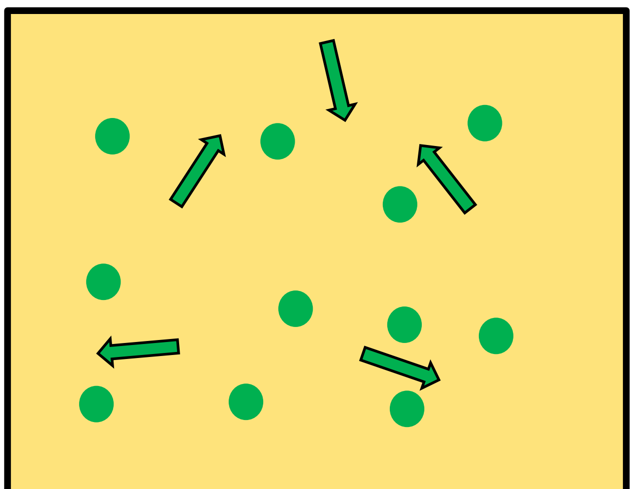


Itinerant (metals): natural description in k-space



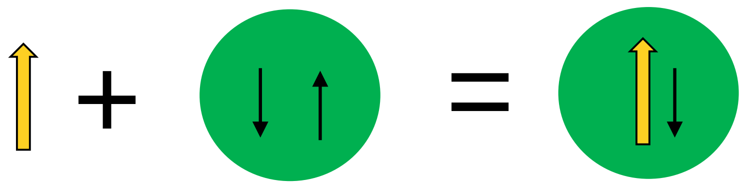
Local (insulators): natural description in real space

Correlated (Kondo): No “natural” language



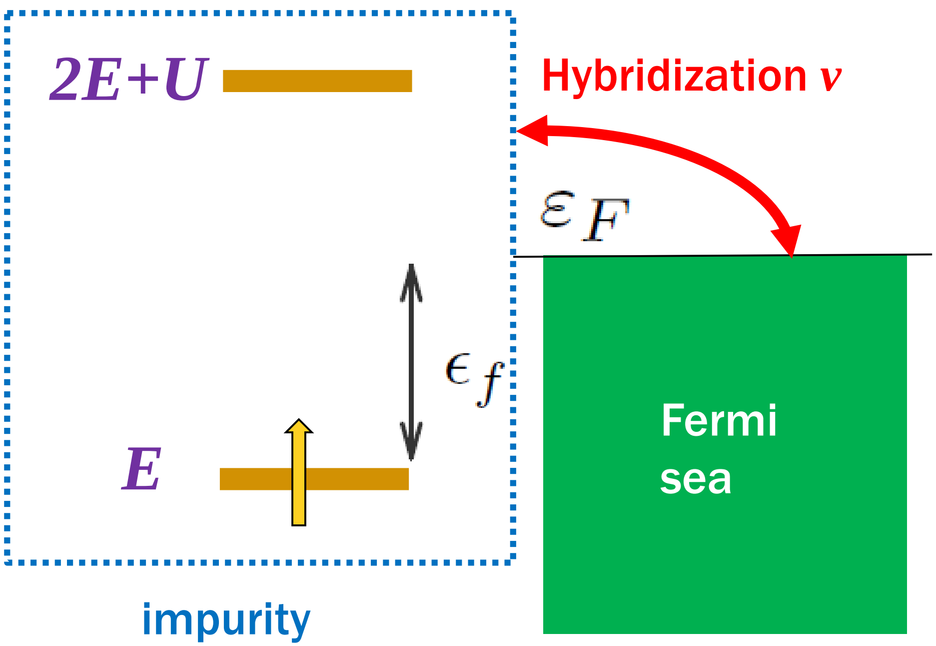
Interactions often “mix representations” and are relevant.

Kondo impurity



screening of impurity spin by conduction electrons

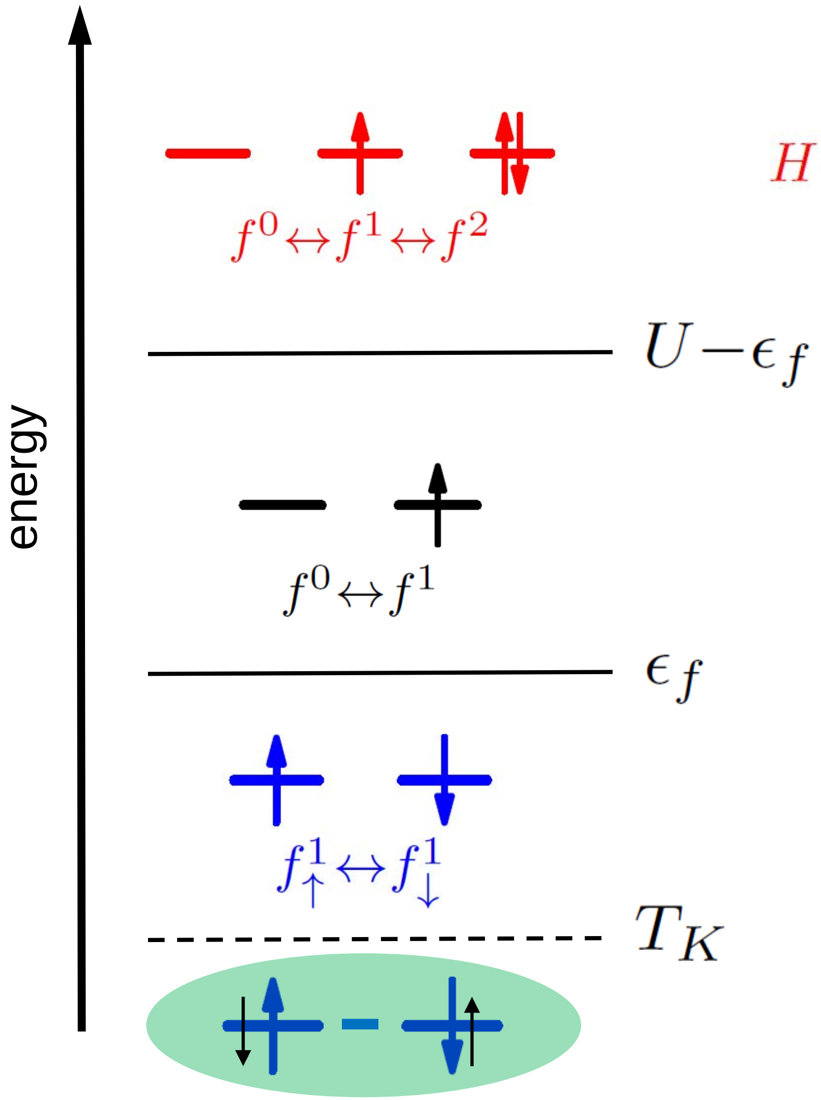
No such thing as pure spin in materials: f-electrons



Anderson impurity model:

Physics depends on the relative magnitude of hybridization vs. location of the f-level vs charging energy U

Single orbital Anderson impurity



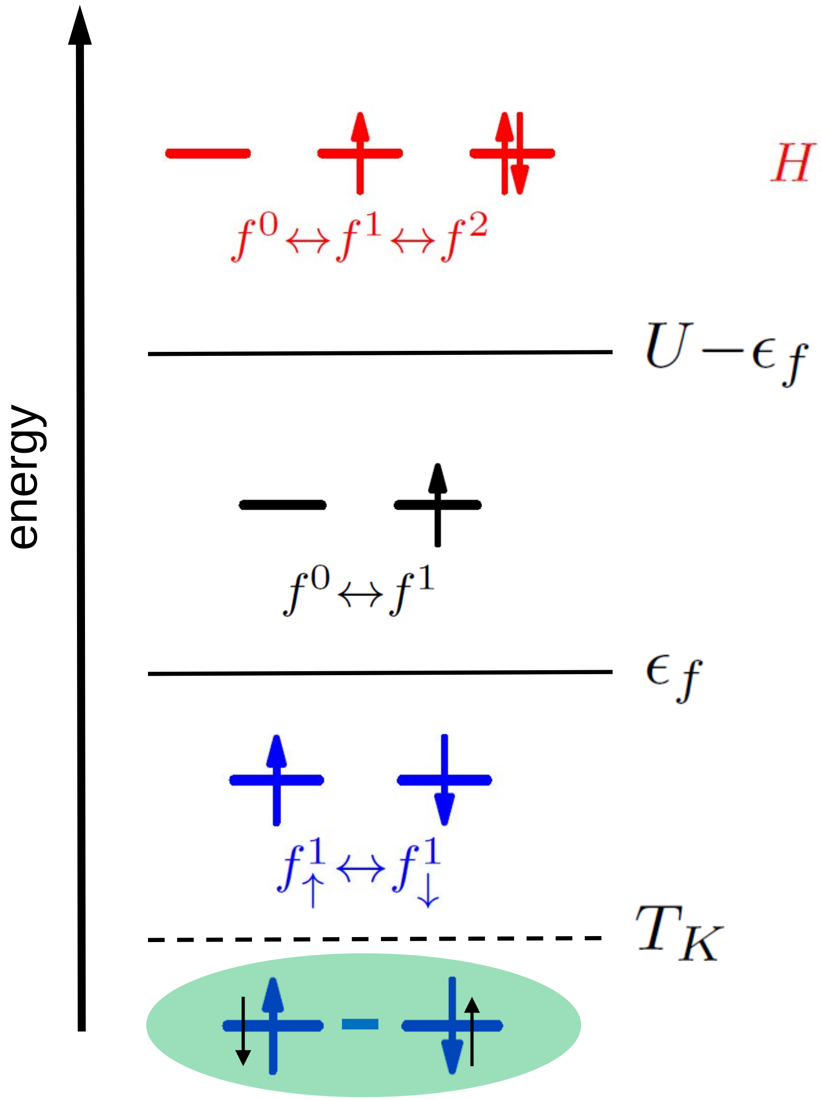
Anderson hamiltonian

$$H = \frac{v}{\sqrt{N}} \sum_{\boldsymbol{k}\sigma} (c^\dagger_{\boldsymbol{k}\sigma} f_\sigma + \text{h.c.}) - \epsilon_f n^f + U n^f_\uparrow n^f_\downarrow$$

Kondo hamiltonian

$$H = J_K \boldsymbol{S} \cdot c^\dagger_{0,\alpha} \boldsymbol{\sigma}_{\alpha\beta} c_{0,\beta}$$

Single orbital Anderson impurity



Anderson hamiltonian

$$H = \frac{v}{\sqrt{N}} \sum_{\boldsymbol{k}\sigma} (c^\dagger_{\boldsymbol{k}\sigma} f_\sigma + \text{h.c.}) - \epsilon_f n^f + U n^f_\uparrow n^f_\downarrow$$

<u>Schrieffer-Wolff transformation</u>

$$e^-_\uparrow + f^1_\downarrow \leftrightarrows f^2 \leftrightarrows e^-_\downarrow + f^1_\uparrow$$
$$h^+_\uparrow + f^1_\downarrow \leftrightarrows f^0 \leftrightarrows h^+_\downarrow + f^1_\uparrow$$

$J_K \sim v^2/U$

Kondo hamiltonian

$$H = J_K \boldsymbol{S} \cdot c^\dagger_{0,\alpha} \boldsymbol{\sigma}_{\alpha\beta} c_{0,\beta}$$

Kondo screening

- Singlet ground state ***S=0***
- Kondo temperature

$$T_K = De^{-1/g_F J_K}$$

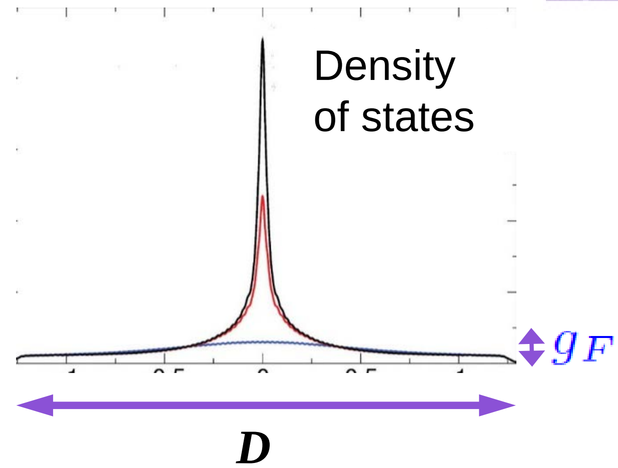
- Kondo peak in the density of states

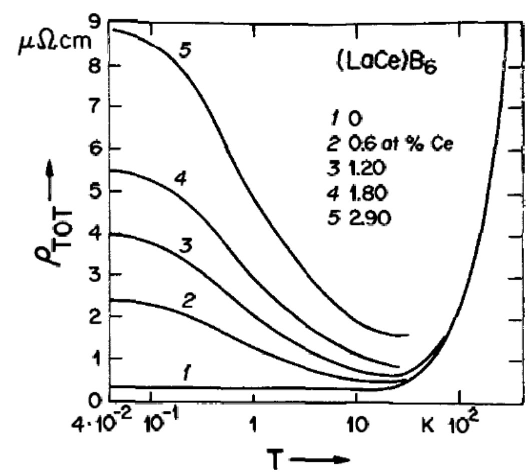
$$g_K(\epsilon) = g_0(\epsilon) + \frac{c_i}{\pi}\frac{\gamma}{\epsilon^2+\gamma^2}$$

- Resisitivity minimum

$$\rho(T) = \rho_0\left[1 - 4J_K g_F \ln T/D\right]$$

- Fermi liquid at low T



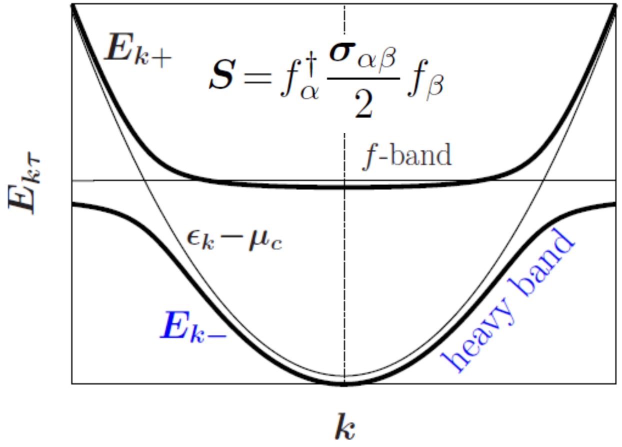


Kondo Lattices and competing orders

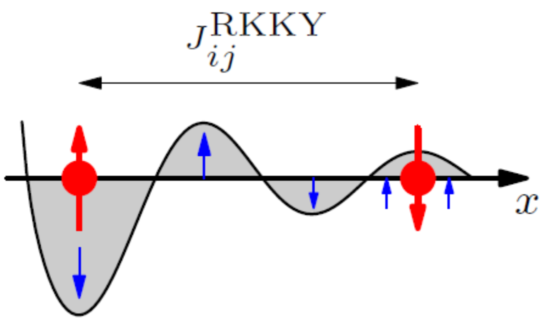
Dense Kondo systems

$$H = \underbrace{\sum_{\boldsymbol{k}\sigma}(\epsilon_{\boldsymbol{k}} - \mu_c)c^\dagger_{\boldsymbol{k}\sigma}c_{\boldsymbol{k}\sigma}}_{\text{conduction band}} + \underbrace{\frac{J_K}{2}\sum_i \boldsymbol{S}_i \cdot c^\dagger_{i\alpha}\boldsymbol{\sigma}_{\alpha\beta}c_{i\beta}}_{\text{Kondo interaction}}$$

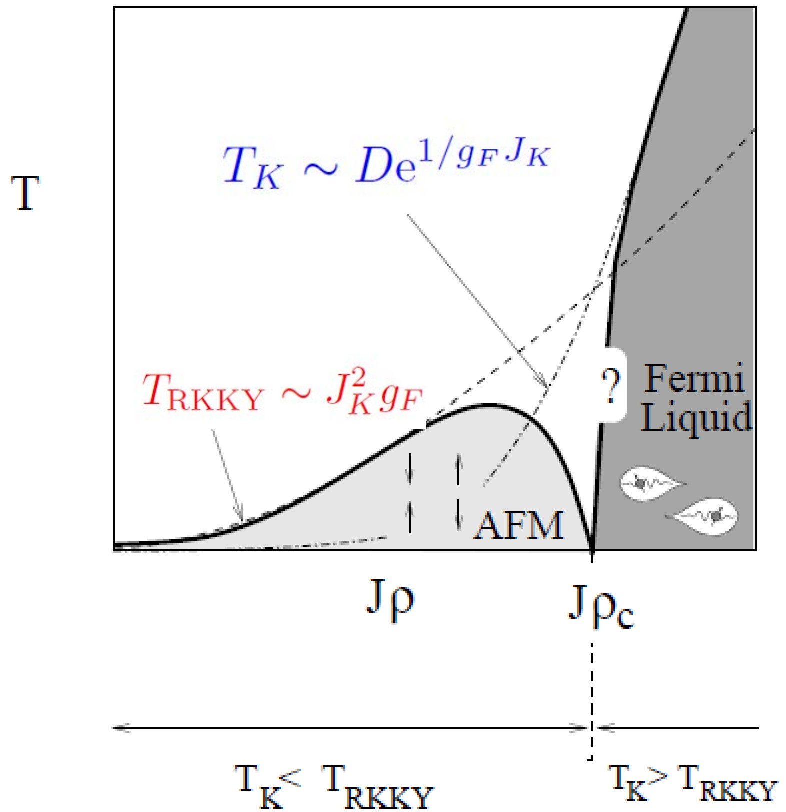
A. Formation of heavy fermion state
   - each of $\boldsymbol{n_f}$ spins screened
   - Fermi surface volume $\boldsymbol{n_f} + \boldsymbol{n_c}$
   - singlet paramagnetic state



B. Formation of the magnetic state
   - no spins screened
   - Exchange (RKKY) interaction
   - Magnetic order of spins



Doniach phase diagram



S. Doniach, 1977

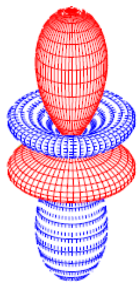
Fig. from P. Coleman, 2007

- **Antagonism of magnetic order and formation of the heavy fermion state**
- **Quantum critical phenomena, emergence of novel phases, superconductivity…**

Real f-electrons

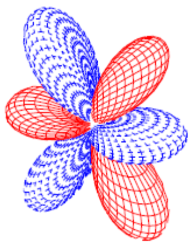
Isolated f-electron atom: 7 different states of orbital quantum numbers

m=0



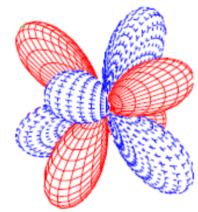
$z(5z^2 - 3r^2)$

m=|1|



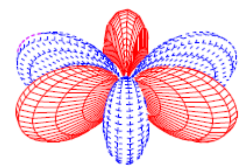
$(5z^2 - r^2)(x \pm \mathrm{i}y)$

m=|2|



$z(x \pm \mathrm{i}y)^2$

m=|3|



$(x \pm \mathrm{i}y)^3$

f-electrons in crystals: degeneracy is lifted due to

$$\left.\begin{array}{l}\text{• Hund's rule coupling}\\ \text{• crystal field splitting}\\ \text{• spin-orbit interaction}\end{array}\right\}$$

- **Hund's rule coupling**
- **crystal field splitting**
- **spin-orbit interaction**

new types of Kondo possible for several electrons in the f-shell

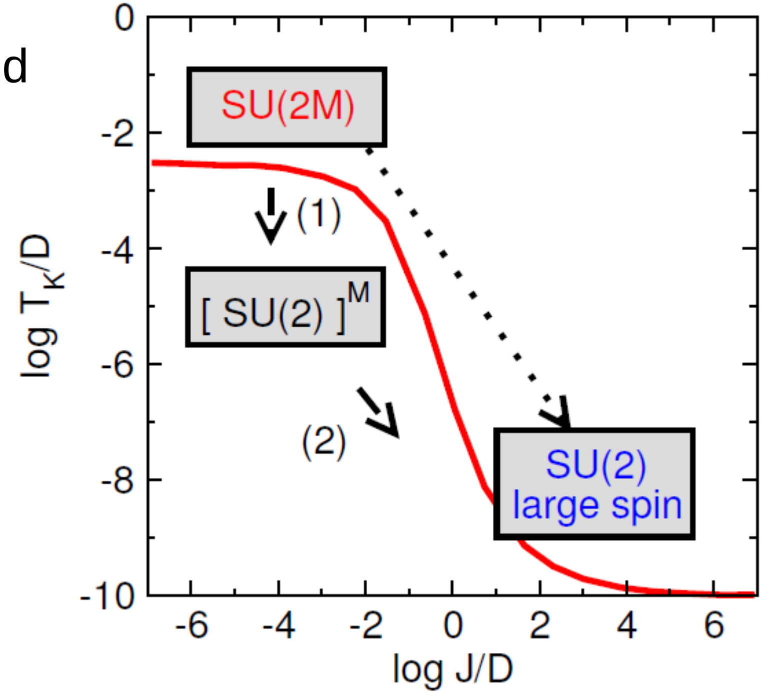
Hierarchy of energy scales is important

Single magnetic impurity

- **free ion** *(Coqblin-Schrieffer model).*
  conduction electrons exchange both spin and orbital moment with the impurity.
  $N_{channels}=N_{imp.levels}$

- ***Hund's rule coupling:***
  freezing of orbital component;
  lifting of orbital degeneracies;
  reduction of Kondo temperature

- ***Kondo coupling:***
  restores local degeneracies;
  frustrates Hund's rule.



A. Georges et al. 2012

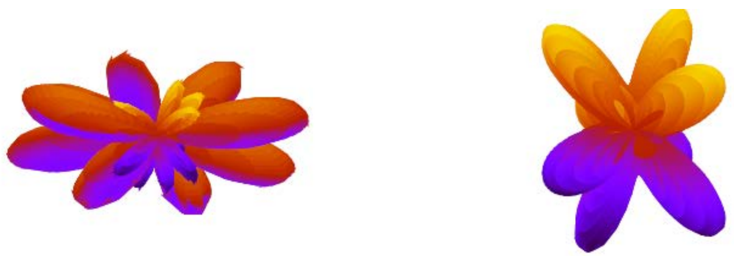
Quadrupolar Kondo and orbital order

D. L. Cox and A. Zawadowski, 2008

U^{4+} ion (5f^2) in cubic environment: Γ_3 pseudospin doublet

$$|+\rangle = \sqrt{\tfrac{7}{24}}\left(|4,4\rangle + |4,-4\rangle\right) - \sqrt{\tfrac{5}{12}}|4,0\rangle; \quad |-\rangle = \tfrac{1}{\sqrt{2}}\left(|4,2\rangle + |4,-2\rangle\right)$$

No magnetic moment, but quadrupolar moment



Coupling to conduction electrons: competition between

- Kondo screened phase
- Orbitally ordered phase due to RKKY quadrupolar interaction

- **Antagonism of orbital order and heavy fermion state**
- **Is it always the case?**

Energy scales

energy

3d	*4f*	*5f*
Hund J	*Hund J*	spin-orbit interaction (SOI)
Crystal electric field (CEF) splitting	spin-orbit interaction (SOI)	Crystal electric field (CEF) splitting
spin-orbit interaction (SOI)	Crystal electric field (CEF) splitting	*Hund J*
		j-j coupling ???

T. Hotta, 2006

Energy scales

energy

$$\boldsymbol{S} = (g_J - 1)\boldsymbol{J} \;\Rightarrow$$

$$-E_{\mathrm{Hund}}\boldsymbol{S}^2 = -(g_j - 1)^2 E_{\mathrm{Hund}}\boldsymbol{J}^2$$

Lande factor g_J-$1<<1$

Effective reduction of the Hund energy scale

5f

spin-orbit interaction (SOI)

Crystal electric field (CEF) splitting

Hund J

j-j coupling ???

T. Hotta, 2006

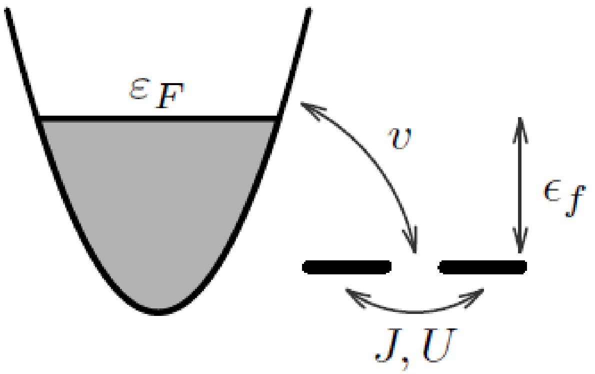
Two-orbital Anderson impurity model

$$H = \underbrace{\sum_{\boldsymbol{k}\sigma} \varepsilon_{\boldsymbol{k}} c^\dagger_{\boldsymbol{k}\sigma} c_{\boldsymbol{k}\sigma}}_{\text{conduction band}} + \underbrace{\frac{v}{\sqrt{N}} \sum_{\boldsymbol{p}\sigma a} (c^\dagger_{\boldsymbol{p}\sigma} f_{a\sigma} + \text{h.c.})}_{\text{Anderson hybridization}} +$$

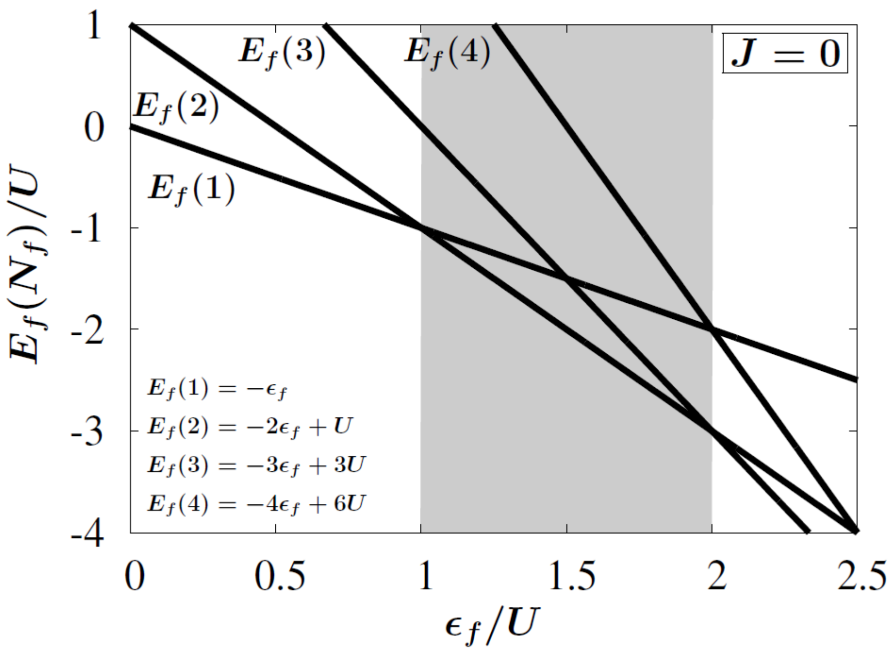
$$+ \underbrace{(J - \epsilon_f) N_f - J\left(\boldsymbol{S}_f^2 + \frac{N_f^2}{4}\right) + \frac{U}{2} N_f (N_f - 1)}_{\text{impurity Hamiltonian}}$$

- *a=1,2:* orbital label
- *J:* Hund's coupling
- *U:* Coulomb repulsion
- Simplifying assumptions

$v_{\boldsymbol{p}a} = v,\; J_{ab} = J,\; U_{ab} = U$



Local two-electron states

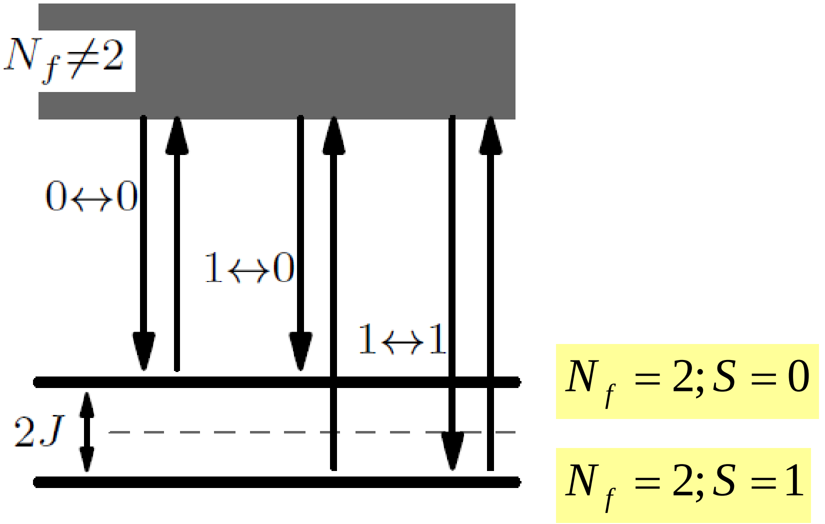


Assumptions

- $J \ll U \sim \epsilon_f$
- f^2 configuration: $\boldsymbol{N_f = 2}$ (six local states)

$\boxed{S_f = 0}$ $\quad E_f = -2\epsilon_f + U + J$	$\boxed{S_f = 1}$, $\quad E_f = -2\epsilon_f + U - J$
$\lvert 00\rangle = \frac{1}{\sqrt{2}}(f^\dagger_{1\uparrow}f^\dagger_{2\downarrow} - f^\dagger_{1\downarrow}f^\dagger_{2\uparrow})\lvert 0_f\rangle$	$\lvert 1,+1\rangle = f^\dagger_{1\uparrow}f^\dagger_{2\uparrow}\lvert 0_f\rangle$
$\lvert s\rangle = \frac{1}{\sqrt{2}}(f^\dagger_{1\uparrow}f^\dagger_{1\downarrow} + f^\dagger_{2\uparrow}f^\dagger_{2\downarrow})\lvert 0_f\rangle$	$\lvert 1,-1\rangle = f^\dagger_{1\downarrow}f^\dagger_{2\downarrow}\lvert 0_f\rangle$
$\lvert a\rangle = \frac{1}{\sqrt{2}}(f^\dagger_{1\uparrow}f^\dagger_{1\downarrow} - f^\dagger_{2\uparrow}f^\dagger_{2\downarrow})\lvert 0_f\rangle$	$\lvert 1,0\rangle = \frac{1}{\sqrt{2}}(f^\dagger_{1\uparrow}f^\dagger_{2\downarrow} + f^\dagger_{1\downarrow}f^\dagger_{2\uparrow})\lvert 0_f\rangle$

Kondo Hamiltonian



Schrieffer-Wolff transformation

$$\widetilde{H} = \mathrm{e}^{\mathcal{S}} H \mathrm{e}^{-\mathcal{S}}$$

$$H_{\mathrm{Kondo}} = \boxed{H_4} \oplus H_2$$

$$H_2 \sim (1 + \sigma^x) n_0^c$$

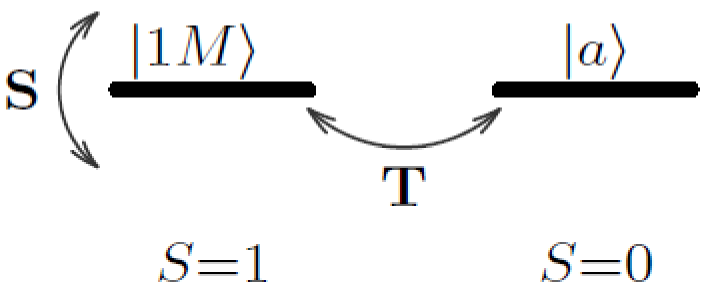
no spin-flips: irrelevant

A quartet of states couples non-trivially to conduction band:

triplet states + antisymmetric singlet

Emergent SO(4) algebra

$$H = \sum_{\boldsymbol{k}\sigma} \varepsilon_{\boldsymbol{k}\sigma} c^{\dagger}_{\boldsymbol{k}\sigma} c_{\boldsymbol{k}\sigma} + J_K \left(\boldsymbol{S} + \boldsymbol{T}\right) \cdot \underbrace{\frac{1}{2} c^{\dagger}_{0\alpha} \boldsymbol{\sigma}_{\alpha\beta} c_{0\beta}}_{\boldsymbol{s}^c_0} + \underbrace{\frac{J}{3}\left(2\boldsymbol{T}^2 - \boldsymbol{S}^2\right)}_{\text{Hund coupling}}$$



Spin generators ($S = 1$)

$$S^+ = \sqrt{2}\left(|10\rangle\langle 1,-1| + |11\rangle\langle 10|\right),$$
$$S^z = |11\rangle\langle 11| - |1,-1\rangle\langle 1,-1|$$

Local algebra

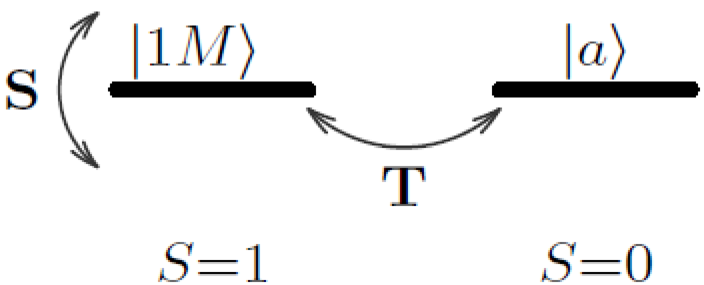
$$[S^i, S^j] = \mathrm{i}\varepsilon_{ijk} S^k$$
$$[T^i, T^j] = \mathrm{i}\varepsilon_{ijk} S^k$$
$$[S^i, T^j] = \mathrm{i}\varepsilon_{ijk} T^k$$

Spin-orbital (Runge-Lenz-like) operator

$$T^+ = \sqrt{2}\left(|11\rangle\langle a| - |a\rangle\langle 1,-1|\right),$$
$$T^z = -\left(|10\rangle\langle a| + |a\rangle\langle 10|\right)$$

Emergent SO(4) algebra

$$H=\sum_{\boldsymbol{k}\sigma}\varepsilon_{\boldsymbol{k}\sigma}c^{\dagger}_{\boldsymbol{k}\sigma}c_{\boldsymbol{k}\sigma}+J_K\underbrace{(\boldsymbol{S}+\boldsymbol{T})}_{\text{pseudospin}}\cdot\underbrace{\frac{1}{2}c^{\dagger}_{0\alpha}\boldsymbol{\sigma}_{\alpha\beta}c_{0\beta}}_{\boldsymbol{s}_0^c}+\underbrace{\frac{J}{3}(2\boldsymbol{T}^2-\boldsymbol{S}^2)}_{\text{Hund coupling}}$$



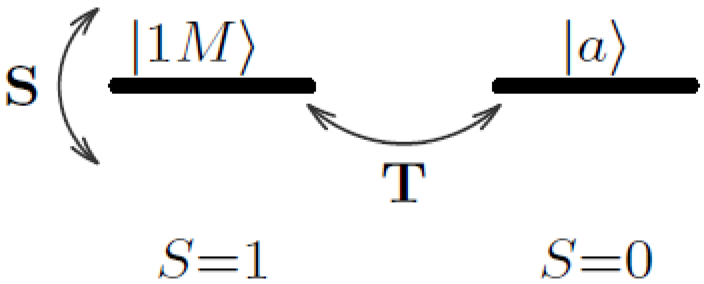
Local algebra
$[S^i,S^j]=\mathrm{i}\varepsilon_{ijk}S^k$
$[T^i,T^j]=\mathrm{i}\varepsilon_{ijk}S^k$
$[S^i,T^j]=\mathrm{i}\varepsilon_{ijk}T^k$

Similar:
SO(4) for Kondo in quantum dots:

- *Y. V. Nazarov and M. Eto, 2000-2001*
- *M. Pustilnik and L. I. Glazman, 2000*
- *K. Kikoin and Y. Avishai, 2001*

Emergent SO(4) algebra

$$H=\sum_{\boldsymbol{k}\sigma}\varepsilon_{\boldsymbol{k}\sigma}c^{\dagger}_{\boldsymbol{k}\sigma}c_{\boldsymbol{k}\sigma}+J_K\underbrace{(\boldsymbol{S}+\boldsymbol{T})}_{\text{pseudospin}}\cdot\underbrace{\frac{1}{2}c^{\dagger}_{0\alpha}\boldsymbol{\sigma}_{\alpha\beta}c_{0\beta}}_{\boldsymbol{s}_0^c}+\underbrace{\frac{J}{3}(2\boldsymbol{T}^2-\boldsymbol{S}^2)}_{\text{Hund coupling}}$$



Local algebra

$$[S^i,S^j]=\mathrm{i}\varepsilon_{ijk}S^k$$
$$[T^i,T^j]=\mathrm{i}\varepsilon_{ijk}S^k$$
$$[S^i,T^j]=\mathrm{i}\varepsilon_{ijk}T^k$$

Orbital pseudospin operator

$$\tau^{\mu}=\frac{1}{2}\sigma^{\mu}_{\alpha\beta}\left(f^{\dagger}_{1\alpha}f_{2\beta}+f^{\dagger}_{2\alpha}f_{1\beta}\right)$$

$$\langle 1M|\boldsymbol{T}|a\rangle=\langle 1M|\boldsymbol{\tau}|a\rangle$$

T-order=orbital order

Kondo Hamiltonian

$$H = \sum_{\boldsymbol{k}\sigma} \varepsilon_{\boldsymbol{k}} c^\dagger_{\boldsymbol{k}\sigma} c_{\boldsymbol{k}\sigma} + J_K \boldsymbol{\Sigma} \cdot c^\dagger_{0\alpha} \boldsymbol{\sigma}_{\alpha\beta} c_{0\beta} \underbrace{- 2J \boldsymbol{\Sigma} \cdot \boldsymbol{A}}_{\text{ferromagnetic coupling}}$$

correlation-driven spin-orbit

- $\boldsymbol{\Sigma} = (\boldsymbol{S} + \boldsymbol{T})/2;\ \boldsymbol{A} = (\boldsymbol{S} - \boldsymbol{T})/2$
- Pseudospin-1/2 degrees of freedom: $\boldsymbol{\Sigma}^2 = \boldsymbol{A}^2 = 3/4$
- $[\Sigma^i, \Sigma^j] = \mathrm{i}\varepsilon_{ijk}\Sigma^k,\ \ [A^i, A^j] = \mathrm{i}\varepsilon_{ijk}A^k,\ \ [\Sigma^i, A^j] = 0$

Kondo lattice Hamiltonian

$$H = \sum_{\boldsymbol{k}\sigma} (\varepsilon_{\boldsymbol{k}} - \mu) c^\dagger_{\boldsymbol{k}\sigma} c_{\boldsymbol{k}\sigma} + 2J_K \sum_i \boldsymbol{\Sigma}_i \cdot \boldsymbol{s}^c_i - 2J \sum_i \boldsymbol{\Sigma}_i \cdot \boldsymbol{A}_i$$

Physical observations:

- Orbital order corresponds to T-ordering

$$\tau^\mu = \frac{1}{2}\sigma^\mu_{\alpha\beta} f^\dagger_{1\alpha} f_{2\beta} + \text{h.c.}; \qquad \langle 1M|\boldsymbol{T}|a\rangle = \langle 1M|\boldsymbol{\tau}|a\rangle$$

- There is no classical orbital order; large Hund → ***S=1 spiral***

$$\boldsymbol{A}_i || \boldsymbol{\Sigma}_i \qquad\qquad \boldsymbol{\Sigma} + \hat{\boldsymbol{A}} = \boldsymbol{S}$$

- Orbital order may exist only for quantum spins

- May result from Kondo competing with Hund

Heavy fermion state

If no Hund's coupling, $J=0$

- pseudofermion mean field

$$\boldsymbol{\Sigma}_i = \frac{1}{2} h^\dagger_{i\alpha} \boldsymbol{\sigma}_{\alpha\beta} h_{i\beta}$$

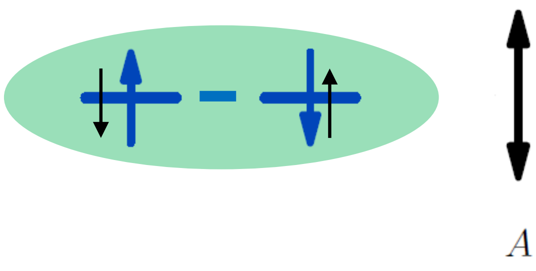
- hybridization order parameter

$$\chi_0 = \frac{1}{\sqrt{2}} \sum_\alpha \langle \mathrm{HF}_{c\Sigma} | c^\dagger_{i\alpha} h_{i\alpha} | \mathrm{HF}_{c\Sigma} \rangle$$

- mean field ground state at $J=0$

c-$\boldsymbol{\Sigma}$ singlets

free $\boldsymbol{A}_i$



Degenerate ground state – need to account for Hund's coupling

Orbital order in the heavy fermion state

Small $J \ll J_K$

heavy fermion state + perturbation

just like RKKY calculation

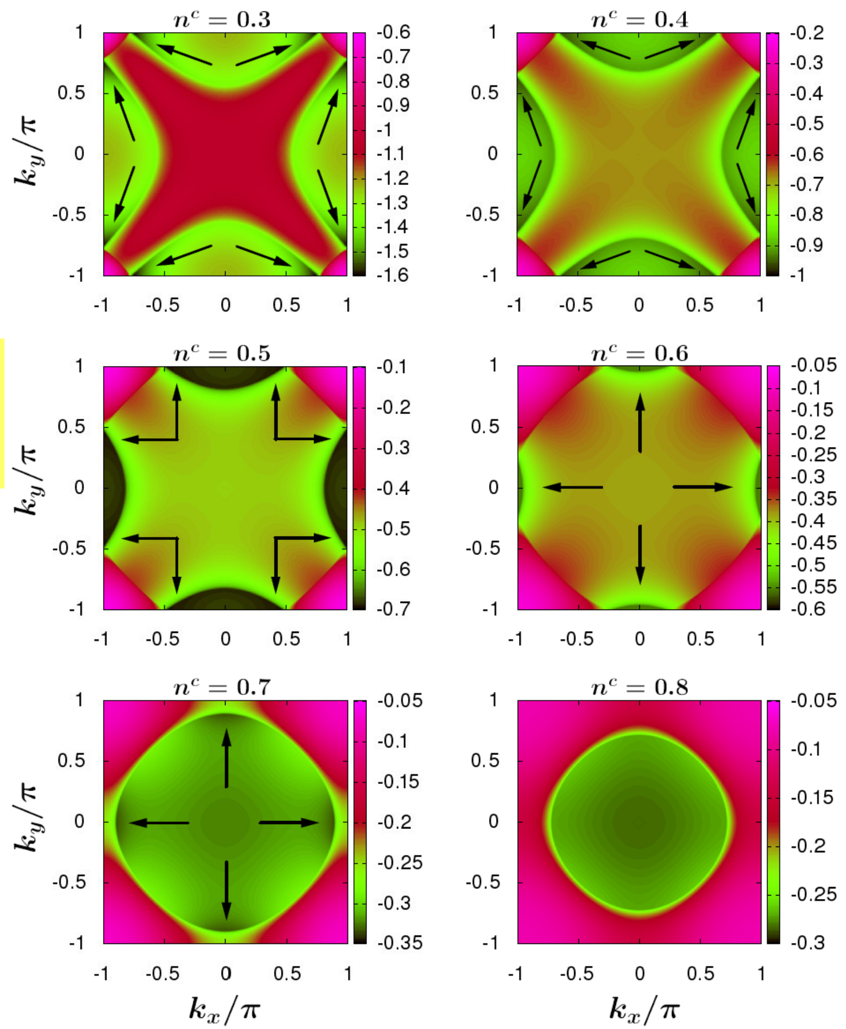
$$H_A^{\mathrm{ef}} = \sum_{ij} I_{ij} \boldsymbol{A}_i \cdot \boldsymbol{A}_j, \;\; I_{ij} \sim J^2/J_K$$

spiral order in $\boldsymbol{A} = (\boldsymbol{S} - \boldsymbol{T})/2$

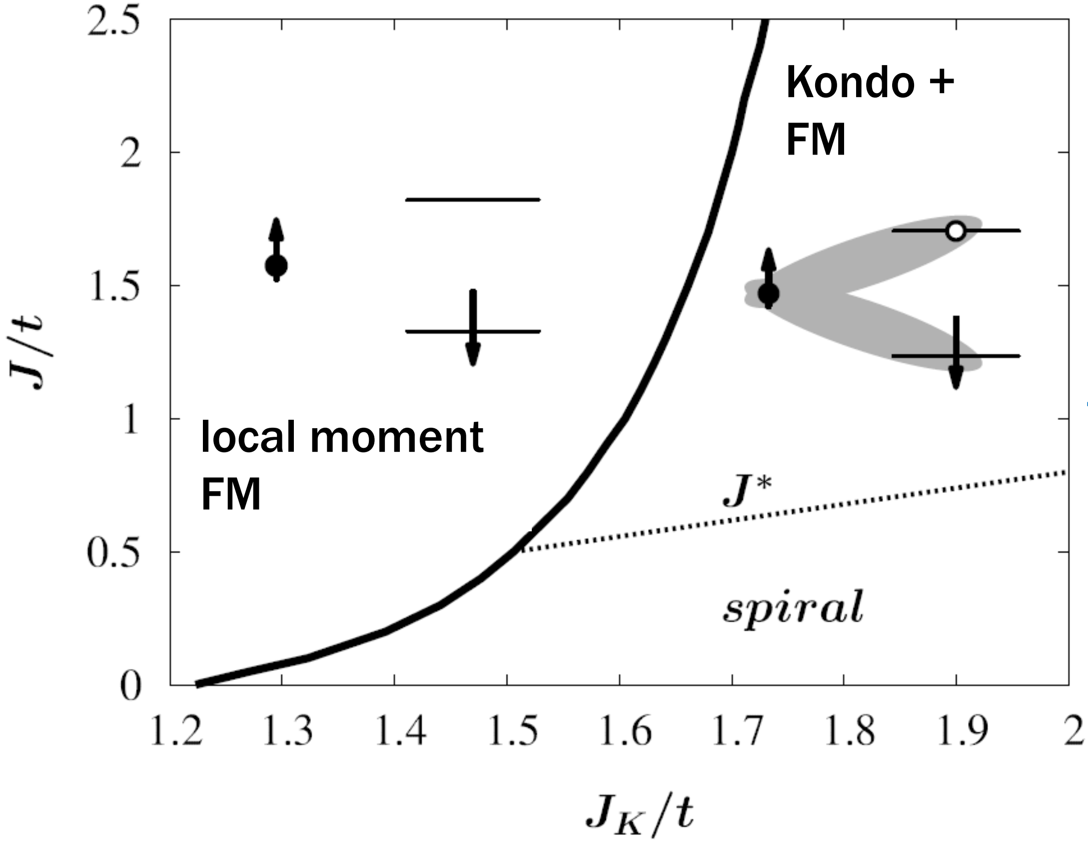
but heavy fermion

$$\langle \psi_0 | \boldsymbol{\Sigma}_i | \psi_0 \rangle = 0 \Rightarrow \langle \boldsymbol{T}_i \rangle = -\langle \boldsymbol{S}_i \rangle$$

Combined spin-orbital spiral state



Phase diagram



transition or crossover depending on symmetry: possible quantum critical line

Conclusions

- Hund's rule is frustrated by the "Kondo" coupling to the orbital degrees or freedom

- The resulting local many-body spin-orbit interaction stabilizes coexistent magnetic and orbital order solely inside the heavy fermion state.

- Neutron and x-ray on 5f (some 4f?) actinide materials? U compounds? some Pr compounds?