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SIMONS FOUNDATION





 I. Non-Fermi liquids in multipolar Kondo systems
 Single multipolar local moment coupled to conduction electrons
 (Focus on cubic Pr³⁺ systems)
 RG and Conformal Field Theory

2. Competition with RKKY and quantum criticality Bose-Fermi Kondo model and quantum criticality

3. Two-stage Kondo destruction and Fermi surface reconstruction: cubic Ce³⁺ systems

Collaborators







Adarsh Patri U of Toronto => MIT SangEun Han U of Toronto Daniel Schulz U of Toronto

A. Patri, YBK, PRX (2020), arXiv:2005.08973
A. Patri, I. Khait, YBK, PR Research (2020), arXiv:1904.02717

Kondo problem

S. Han, D. Schultz, YBK, arXiv:2206.02808 (2022) S. Han, D. Schultz, YBK, arXiv:2207.07661 (2022)

Kondo + RKKY

Multipolar local moments

Many f-electron localized ions carry multipolar moments



Multipolar local moments

Many f-electron localized ions carry multipolar moments



Non-Kramers Doublet: Pr^{3+} $|+\rangle \equiv \Gamma_3^{(1)}$ $|-\rangle \equiv \Gamma_3^{(2)}$

 $\langle \pm | J_{\alpha} | \pm \rangle = 0$ No dipole moment !

But, finite matrix elements for

 $O_{22} = \frac{\sqrt{3}}{2} (J_x^2 - J_y^2) \quad O_{20} = \frac{1}{2} (3J_z^2 - J^2) \quad \text{Quadrupolar}$ $T_{xyz} = \frac{\sqrt{15}}{6} \overline{J_x J_y J_z} \quad \text{Octupolar}$

Pseudo-spin basis
$$S^x = -\frac{1}{4}\mathcal{O}_{22}$$
 $|\uparrow\rangle \equiv \frac{1}{\sqrt{2}}(|\Gamma_3^{(1)}\rangle + i|\Gamma_3^{(2)}\rangle)$ $S^y = -\frac{1}{4}\mathcal{O}_{20}$ $|\downarrow\rangle \equiv \frac{1}{\sqrt{2}}(i|\Gamma_3^{(1)}\rangle + |\Gamma_3^{(2)}\rangle)$ $S^z = \frac{1}{3\sqrt{5}}\mathcal{T}_{xyz}$ (S^x, S^y) Quadrupolar S^z Octupolartime-reversal eventime-reversal odd



Non-Kramers Doublet: Pr^{3+} $|+\rangle \equiv \Gamma_3^{(1)}$ $|-\rangle \equiv \Gamma_3^{(2)}$

Unusual forms of the Kondo and RKKY interaction

First, look at the single impurity Kondo problem !

Pseudo-spin basis $S^x = -\frac{1}{4}\mathcal{O}_{22}$ $|\uparrow\rangle \equiv \frac{1}{\sqrt{2}}(|\Gamma_3^{(1)}\rangle + i|\Gamma_3^{(2)}\rangle)$ $S^y = -\frac{1}{4}\mathcal{O}_{20}$ $|\downarrow\rangle \equiv \frac{1}{\sqrt{2}}(i|\Gamma_3^{(1)}\rangle + |\Gamma_3^{(2)}\rangle)$ $S^z = \frac{1}{3\sqrt{5}}\mathcal{T}_{xyz}$ (S^x, S^y) Quadrupolar S^z Octupolartime-reversal eventime-reversal odd



PHYSICAL REVIEW LETTERS 121, 077206 (2018)

Single-Site Non-Fermi-Liquid Behaviors in a Diluted $4f^2$ System $Y_{1-x}Pr_xIr_2Zn_{20}$

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How to model conduction electrons ?

Local symmetry around Pr ion is T_d Fermi pockets around zone center (quantum oscillations, S. Nagashima et al, 2014) Conduction electron orbitals can be classified in terms of irreducible representation of T_d

 $\begin{array}{c|c} \mathbf{A_1} & xyz, & x^4 + y^4 + z^4, & x^2y^2z^2 \\ \hline \mathbf{A_2} & x^4(y^2 - z^2) + y^4(z^2 - x^2) + z^4(x^2 - y^2) \\ \hline \mathbf{E} & \{x^2 - y^2, & 2z^2 - x^2 - y^2\} \\ \hline \mathbf{T_1} & \{x(z^2 - y^2), & y(z^2 - x^2), & z(x^2 - y^2)\} \\ \hline \mathbf{T_2} & \{x, & y, & z\}, & \{xy, & xz, & yz\}, \end{array}$

For concrete models, we consider T₂ (p- or t_{2g} d-orbitals) - "molecular" orbitals

The "Kondo" model

 S^x, S^y, S^z couple to fermion spin-orbital 'currents' that transform exactly in the same way

For example,

$$\begin{aligned} H_{3} &= K_{3}S^{z} \begin{bmatrix} \sigma_{\alpha\beta}^{x}(c_{p_{y},\alpha}^{\dagger}c_{p_{z},\beta}+h.c.) & \alpha,\beta =\uparrow,\downarrow \\ &+ \sigma_{\alpha\beta}^{y}(c_{p_{z},\alpha}^{\dagger}c_{p_{x},\beta}+h.c.) & three \\ &+ \sigma_{\alpha\beta}^{z}(c_{p_{x},\alpha}^{\dagger}c_{p_{y},\beta}+h.c.) \end{bmatrix} & \alpha,\beta =\uparrow,\downarrow \\ & three \\ K_{1},K_{2},K_{3} \text{ independent } \\ & couplings \end{aligned}$$

Entangled fluctuations of both orbital and spin ! Perform RG computations up to 3rd order: two stable fixed points

A. Patri, I. Khait, YBK, PR Research 2, 013257 (2020)

The "Kondo" model

 S^x, S^y, S^z couple to fermion spin-orbital 'currents' that transform exactly in the same way

For example,

L=1, S=1/2
$$|j, j_z\rangle = |j = 3/2, j_z\rangle, |j = 1/2, j_z\rangle$$
 $|p_x, \uparrow\rangle$ $|p_x, \downarrow\rangle$ Spin-orbital
entangled
basis $|3/2, 3/2\rangle$ $|3/2, 1/2\rangle$ $|j, j_z\rangle = |j = 3/2, j_z\rangle, |j = 1/2, j_z\rangle$ $|3/2, 3/2\rangle$ $|3/2, -1/2\rangle$ $|p_z, \uparrow\rangle$ $|p_z, \downarrow\rangle$ $|1/2, -1/2\rangle$ $|1/2, -1/2\rangle$

Fixed point I Two channel Kondo model ! Only j=3/2 $e_2^3 e_2^3$ from are in v_2^3 by d S^x $S^{w} = \frac{1}{1 + 1} \frac{3}{2} \frac{3}{2} \frac{3}{2} \frac{1}{2} \frac{3}{2} \frac{3}{2} \frac{1}{2} \frac{3}{2} \frac{3}{2}$ $\begin{vmatrix} \mathbf{\hat{S}}^{y-1} \\ 2^{,} \overline{2} \end{vmatrix} \begin{array}{c} C_v \propto T \ln T \\ C_v \propto \sqrt{T} \ln T \\ \frac{1}{2} \begin{vmatrix} \frac{1}{2} \end{vmatrix} \\ \rho \propto \sqrt{T} \end{aligned}$ S^x

 S^x

 S^x

 S^z

 S^x

Fixed point II

33222

 $\frac{-1}{\overline{2}^{1}}$

 $\begin{pmatrix} 1\\ \frac{1}{2}\\ 2 \end{pmatrix}$

 $\frac{-2^3}{2}$

All of j=3/2 and j=1/2 electrons are involved



Two channels

But each channel involves three flavors of conduction electrons - SU(3) 'current'

This is not the usual Kondo problem

A. Patri, YBK, PRX, arXiv:2005.08973 (2020)



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Approaching the fixed point by fixing the ratios of the couplings

$$g_k^* = 1$$
 at the fixed point

dimension of the leading irrelevant operator $1 + \Delta$ $\Delta = 1/4$ $\rho \propto T^{\Delta} = T^{1/4}$ $C_v \propto T^{2\Delta} = T^{1/2}$

Fixed point II

True nature of this new fixed point ? Rewriting the Kondo coupling

$$H_k = g_k \sum_{m=1,2} \vec{\psi}_m^{\dagger}(0) \left[\frac{S^x}{2} \frac{\lambda^4}{2} + \frac{S^y}{2} \frac{\lambda^6}{2} + \frac{S^z}{2} \frac{\lambda^2}{2} \right] \vec{\psi}_m(0)$$

 λ^a a = 1, ..., 8 **3x3** SU(3) Gell-Mann matrices

$$\begin{split} \vec{\psi}_{m=1}^{\dagger} &= \left(-c_{\frac{3}{2},\frac{3}{2}}^{\dagger}, -c_{\frac{3}{2},\frac{-1}{2}}^{\dagger}, c_{\frac{1}{2},\frac{-1}{2}}^{\dagger} \right) & g_{k}^{*} = 1 \\ \vec{\psi}_{m=2}^{\dagger} &= \left(c_{\frac{3}{2},-\frac{3}{2}}^{\dagger}, c_{\frac{3}{2},\frac{1}{2}}^{\dagger}, c_{\frac{1}{2},\frac{1}{2}}^{\dagger} \right) & \text{at the perturbative} \\ \mathbf{fixed point} \end{split}$$

This is not an SU(3) Kondo problem ...

Current Algebra and Conformal Field Theory (Results)

Useful to generalize this to k channels Here k = 2

$$g_k^* = \frac{2}{k+3} \qquad \Delta = \frac{2}{4k+2}$$

 $1+\Delta$ $\,$ dimension of the leading irrelevant operator $\,$

 $\begin{array}{ccc} C_v \propto T^{2\Delta} & \rho \propto T^{\Delta} \\ \hline \mathbf{Perturbative regime} & k \gg 1 \\ g_k^* \rightarrow 2/k & \Delta \rightarrow 1/2k \\ k = 2 & g_k^* \rightarrow 1 & \Delta \rightarrow 1/4 \\ \hline \mathbf{recover the perturbative results !} \end{array} \qquad \begin{array}{c} \mathbf{Exact} & k = 2 \\ g_k^* = 2/5 \\ \Delta = 1/5 \\ C_v/T \propto T^{-3/5} \\ \rho \propto T^{1/5} \end{array}$

A. Patri, YBK, PRX, arXiv:2005.08973 (2020)

Current Algebra and Conformal Field Theory Solution (Schematic)

Consider δ -function interaction and s-wave scattering Mapping to I+I dimensional (chiral) model



Current Algebra and Conformal Field Theory Solution (Schematic)

Bulk free theory (before coupling to impurity)

Current algebra and conformal embedding

 $\mathrm{U}(1) \times \mathrm{SU}(3)_2 \times \mathrm{SU}(2)_3$

Charge Spin-Orbital Flavor(channel)

3 = three spin-orbital bands in each channel2 = two channels

$$H_{0} = \frac{1}{12} : JJ : (z) + \frac{1}{5} : J^{a}J^{a} : (z) + \frac{1}{5} : J^{A}J^{A} : (z)$$

Charge Spin-Orbital Flavor(channel)
 $a = 1, ..., 8$ $A = 1, ..., 3$

A. Patri, YBK, arXiv:2005.08973 (2020)

Current Algebra and Conformal Field Theory Solution (Schematic)

Bulk free theory (before coupling to impurity)

Current algebra and conformal embedding

 $U(1) \times SU(3)_2 \times SU(2)_3$

Charge Spin-Orbital Flavor(channel)

But only three of eight generators of SU(3) are coupled to the impurity

$$H_K = g_k \left(J^4(z) \frac{S^x}{2} + J^6(z) \frac{S^y}{2} + J^2(z) \frac{S^z}{2} \right)$$
 Kondo coupling

We need a different conformal embedding ...

A. Patri, YBK, arXiv:2005.08973 (2020)

Current Algebra and Conformal Field Theory Solution (Schematic)

Bulk free theory $U(1) \times SU(3)_2 \times SU(2)_3$

Charge Spin-Orbital Flavor(channel) Conformal embedding

Coset construction

 $SU(3)_2 = [3-state Potts model] \times SU(2)_8$ $J^2, J^4, J^6 \in \widetilde{SU}(2)_8$

Kondo coupling imposes (conformally invariant) boundary conditions on the currents

Boundary CFT (John Cardy) Affleck + Ludwig (1991)

Rearrangement of the conformal towers

Experiments ?



The fits are encouraging ! We need sharper predictions ...

Competition with RKKY: Bose-Fermi Kondo model

$$H = H_{\text{Fermi Kondo}} + H_{\text{Bose Kondo}}$$

$$H_{\text{RKKY}} = \sum_{ij} \left[J_{ij}^{Q} (S_{i}^{x} S_{j}^{x} + S_{i}^{y} S_{j}^{y}) + J_{ij}^{O} S_{i}^{z} S_{j}^{z} \right]$$
$$\longrightarrow H_{\text{Bose Kondo}} = g_{Q} (S^{x} \phi_{0}^{x} + S^{y} \phi_{0}^{y}) + g_{O} S^{z} \phi_{0}^{z}$$
$$+ \sum_{\mathbf{k}} \left[\Omega_{Q\mathbf{k}} (\phi_{\mathbf{k}}^{x\dagger} \phi_{\mathbf{k}}^{x} + \phi_{\mathbf{k}}^{y\dagger} \phi_{\mathbf{k}}^{y}) + \Omega_{O\mathbf{k}} \phi_{\mathbf{k}}^{z\dagger} \phi_{\mathbf{k}}^{z} \right]$$

 $\begin{array}{ll} \mbox{Boson} \\ \mbox{density of states} \end{array} \quad D_i(\omega) = C_i |\omega|^{1-\epsilon_i} {\rm sgn}(\omega) \qquad i = Q, O \end{array}$

This is a local approximation of the RKKY interaction in the spirit of DMFT

J. L. Smith, Q. Si (96,97), A. M. Sengupta (97)

S. Han, D. Schultz, YBK, arXiv:2206.02808 (2022)

RKKY Quadrupolar Ordering (Small FS)







S. Han, D. Schultz, YBK, arXiv:2206.02808 (2022)

RKKY Quadrupolar Ordering (Small FS)



S. Han, D. Schultz, YBK, arXiv:2206.02808 (2022)

RKKY Quadrupolar Ordering (Small FS)



Quadrupolar susceptibility $\chi^Q(\tau) = \langle T_\tau S^{x,y}(\tau) S^{x,y}(0) \rangle \propto \left(\frac{\tau_0}{|\tau|}\right)^{\gamma_Q}$ Octupolar susceptibility $\chi^O(\tau) = \langle T_\tau S^z(\tau) S^z(0) \rangle \propto \left(\frac{\tau_0}{|\tau|}\right)^{\gamma_Q}$

RKKY Quadrupolar Ordering (Small FS)



Measurement via elastic constants (ultrasound)

$$(C_{11} - C_{12}) = (C_{11}^0 - C_{12}^0) - (s_Q^2)\chi'_Q,$$

$$C_{44} = C_{44}^0 - (s_Q^2 h^2)\chi'_O,$$

RKKY Quadrupolar Ordering (Small FS)



Measurement via elastic constants (ultrasound)

$$(C_{11} - C_{12}) = (C_{11}^0 - C_{12}^0) - (s_Q^2)\chi'_Q,$$

$$C_{44} = C_{44}^0 - (s_Q^2 h^2)\chi'_O,$$

Multipolar local moments with two-degenerate Kramers doublets (Quartet)

Ce³⁺ [Xe]4f¹ J=5/2

Ce₃Pd₂₀Si₆ S. Paschen et al (2008) Ce₃Pd₂₀Ge₆ J.Kitagawa et al (1996)

In cubic systems, two lowest Kramers doublet are degenerate, which makes 4-fold degenerate F₈ CEF level



×

Crystal field splitting



I5 multipolar moments ←→→ I5 generators of SU(4)

| Moments | irreducible representations | Operators |
|---------------------|-----------------------------|---|
| Dipolar moments | T_1 | J_x |
| | | J_y |
| | | J_z |
| Quadrupolar moments | E | $O_2^2 \equiv rac{\sqrt{3}}{2} (J_x^2 - J_y^2)$ |
| | | $O_2^0 \equiv rac{1}{2}(2J_z^2 - J_x^2 - J_y^2)$ |
| | T_2 | $O_{yz} \equiv rac{\sqrt{3}}{2} \overline{J_y J_z}$ |
| | | $O_{zx} \equiv rac{\sqrt{3}}{2} \overline{J_z J_x}$ |
| | | $O_{xy} \equiv \frac{\sqrt{3}}{2} \overline{J_x J_y}$ |
| Octupolar moments | A_2 | $T_{xyz} \equiv \frac{\sqrt{15}}{6} \overline{J_x J_y J_z}$ |
| | T_1 | $T_x^{\alpha} \equiv \frac{1}{2} \left(2J_x^3 - \overline{J_x J_y^2} - \overline{J_z^2 J_x} \right)$ |
| | | $T_y^{\alpha} \equiv \frac{1}{2} \left(2J_y^3 - \overline{J_y J_z^2} - \overline{J_x^2 J_y} \right)$ |
| | | $T_z^{\alpha} \equiv \frac{1}{2} \left(2J_z^3 - J_z J_x^2 - J_y^2 J_z \right)$ |
| | T_2 | $T_x^{\beta} \equiv rac{\sqrt{15}}{6} (\overline{J_x J_y^2} - \overline{J_z^2 J_x})$ |
| | | $T_y^{\beta} \equiv \frac{\sqrt{15}}{6} (\overline{J_y J_z^2} - \overline{J_x^2 J_y})$ |
| | | $T_z^{\beta} \equiv \frac{\sqrt{15}}{6} (\overline{J_z J_x^2} - \overline{J_y^2 J_z})$ |



Bose-Fermi Kondo model (results)



Kondo fixed point

RKKY fixed point

S. Han, D. Schultz, YBK, arXiv:2207.07661 (2022)

Bose-Fermi Kondo model (results)

Hybridization



S. Han, D. Schultz, YBK, arXiv:2207.07661 (2022)

Bose-Fermi Kondo model (results)

Hybridization $\{J_x^2 - J_y^2, 3J_z^2 - J^2\}$ Ordering $\{-21J_{x,y,z}+2\mathcal{T}^{\alpha}_{x,y,z}\}$ $\{J_x^2 - J_y^2, 3J_z^2 - J^2\} \\ \{-21J_{x,y,z} + 2\mathcal{T}_{x,y,z}^{\alpha}\}$ $\{J_x J_y J_z\}$ F'_1 C_{BP} P B_M $\{J_x^2 - J_y^2, 3J_z^2 - \pmb{J}^2\}$ Ordering $\{-21J_{x,y,z}+2\mathcal{T}_{x,y,z}^{\alpha}\}$ Hybridization $\{J_x J_y J_z\}$ **Two-stage** c + D&O + Q + Oc + D&OС Kondo destruction S. Han, D. Schultz, YBK, arXiv:2207.07661 (2022) +Q and D&O +Q



| Irrep. | Notation | In terms of Stevens | Moment |
|----------|----------|---|--------|
| T_{1a} | S^1 | $-\frac{1}{15}J_x + \frac{7}{90}\mathcal{T}_x^{\alpha}$ | D&O |
| T_{1a} | S^2 | $-rac{1}{15}J_y+rac{7}{90}\mathcal{T}_y^lpha$ | D&O |
| T_{1a} | S^3 | $-\frac{1}{15}J_z + \frac{7}{90}\mathcal{T}_z^{lpha}$ | D&O |
| | S^4 | $rac{1}{8}\mathcal{O}_{22}$ | Q |
| | S^5 | $\frac{1}{8}\mathcal{O}_{20}$ | Q |
| T_{2+} | S^6 | $rac{1}{2}\mathcal{O}_{yz}$ | Q |
| T_{2+} | S^7 | $rac{1}{2}\mathcal{O}_{zx}$ | Q |
| T_{2+} | S^8 | $\frac{1}{2}\mathcal{O}_{xy}$ | Q |
| A_2 | S^9 | $rac{1}{9\sqrt{5}}\mathcal{T}_{xyz}$ | О |
| T_{1b} | S^{10} | $-\frac{7}{15}J_x + \frac{2}{45}\mathcal{T}_x^{\alpha}$ | D&O |
| T_{1b} | S^{11} | $-rac{7}{15}J_y+rac{2}{45}\mathcal{T}_y^lpha$ | D&O |
| T_{1b} | S^{12} | $-\frac{7}{15}J_z + \frac{2}{45}\mathcal{T}_z^{\alpha}$ | D&O |
| T_{2-} | S^{13} | $rac{1}{6\sqrt{5}}\mathcal{T}_x^eta$ | Ο |
| T_{2-} | S^{14} | $rac{1}{6\sqrt{5}}\mathcal{T}_y^eta$ | Ο |
| T_{2-} | S^{15} | $rac{1}{6\sqrt{5}}\mathcal{T}_z^eta$ | 0 |

| Irrep. | Moments |
|--------------------|---------------------------------------|
| T_{1a} | $\boxed{S^1,S^2,S^3}$ |
| $\mid E \mid$ | S^4, S^5 |
| $ T_{2+} $ | S^6, S^7, S^8 |
| A_2 | S^9 |
| $\mid T_{1b} \mid$ | $\left S^{10}, S^{11}, S^{12}\right $ |
| T_{2-} | $\left S^{13}, S^{14}, S^{15} ight $ |

Multipolar susceptibility Ultrasound

$$(C_{11} - C_{12}) = (C_{11}^0 - C_{12}^0) - (s_E^2)\chi'_E - 2(s_{2-}^2 h_z^2)\chi'_{2-},$$
(15)

$$C_{44} = C_{44}^0 - (s_{2+}^2)\chi'_{2+} - (s_A^2 h_z^2)\chi'_A, \qquad (16)$$

Summary and Outlook

Non-Fermi liquids in multipolar Kondo problems Possibility of plethora of non-Fermi liquids D.Schultz, A.Patri, YBK, PRR (2021), arXiv:2010.04731 D.Schultz, A.Patri, YBK, PRB (2021), arXiv:2104.11245

Kondo and RKKY: Bose-Fermi Kondo models

Multi-stage Fermi surface reconstruction and quantum critical behaviors (Ce-based system)

Kondo lattice problem ? New opportunities for quantum criticality ? New opportunities for unconventional superconductivity ?