A Hund’s Rule Stabilized Nematic Magnetic State in URu$_2$Si$_2$

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Outline

• Experiments on URu$_2$Si$_2$

• The Under-Compensated Anderson Lattice Model

• A Bare Band Structure

• The spin-rotationally invariant Coulomb interactions

• The Order Parameter

• A Mean-field Approximation

• Magnetic Nematicity

• Conclusions
The Experimental Manifestations of Hidden Order in URu$_2$Si$_2$

- The discovery of superconductivity at $T=1.5$ K and a phase transition at $T=17.5$ K with a large specific heat jump in the heavy-fermion material URu$_2$Si$_2$
- $\gamma = 155$ mJ/mole K$^2$

Maple et al. (1986)

Palstra et al. (1985)
Experimental Manifestations

Maple et al. (1986)

Resistivity & Specific Heat

- Gap of \( \sim 10 \text{ meV} \)
- 40% of the Fermi-Surface gapped
Direct Observation of Gaps

- Far Infrared Spectroscopy (Reflectivity)
  Bonn, Garret and Timusk (1988)

Gaps of 5 and 7.6 meV

$$2\Delta/k_B T_{HO} = 3.6 - 5.1$$
Broken Spin-Rotational Invariance

- **Magnetic Torque**
  Okazaki et al (2011)

- **Shubnikov-de Haas**
  Altarawneh et al (2011)

Figure 3. (a) Schematic configuration of the magnetic torque measurements for ab-plane field rotation by using the micro-cantilever technique. The magnetic field $H$ (blue arrow) induces the magnetization $M$ (green arrow) in the URu$_2$Si$_2$ crystal. The torque along the c axis (red arrow) can be detected by the change in the piezo resistor, which is measured by the bridge configuration. (b) Upper panels show raw magnetic torque curves as a function of the azimuthal angle $\phi$ at several temperatures. All data are measured at $\mu_0H = 4$ T. Middle and lower panels show twofold $\tau_{2\phi}$ and fourfold $\tau_{4\phi}$ components of the torque curves which are obtained from the Fourier analysis.

FIG. 3 (color online). (a) Schematic showing band polarization caused by Zeeman splitting, resulting in the depopulation of the minority spin component above $H_p$, defined in Eq (1). (b) Polar plot of the measured $\theta$-dependent effective $g$ factor in URu$_2$Si$_2$ [19,27] (black symbols) together with a fit to $g^* = g_z \cos \theta$ (black line), where $g_z = 2.6$ (assuming $\frac{1}{2}$ pseudospins), and its comparison with an isotropic $g = 2$ (red line). (c) Schematic of the field-dependent cross-sectional areas of the up-spin and down-spin components for a single pocket, together with the “back projected” quantum oscillation frequency before $F$ and after $F + \Delta F$ polarization. (d) The same schematic in which the frequency shift $\Delta F'$ is reduced by additional pockets acting as a charge reservoir.
The Compensated Anderson Lattice

- N-fold degenerate localized 5f atomic levels $E_f$
- N-fold degenerate conduction band $e_k$

- Hybridization $V_{fd}$

$\rightarrow$ N Hybridized Bands

- Coulomb Interaction $U$ and Hund’s rule Exchange $J$
  between 5f electrons on the same atom

$\rightarrow$ Kondo Effect with a zero magnetic moment

Localized 5f spin $S^z=N/2$ screened by a compensating cloud of conduction electrons with spin $S^z=-N/2$ (forming a spin singlet)
The Under-Compensated Anderson Lattice

- N-fold degenerate semi-localized 5f bands (small direct hopping)
- 1 (spin-only degenerate) non-degenerate itinerant conduction band

- Hybridization $V_{fd}$

→ Hybridized Band and (N-1) Unhybridized bands

- Coulomb Interaction U and Hund’s rule Exchange J between the 5f electrons on the same atom (forms a net atomic spin)

→ Kondo Effect but also yields a net moment of (N-1)/2

Nozieres and Blandin (1980)

Uranium Monochalcogenides and Pnictides
Kondo Effect and Magnetic Ordering
The Bare Bands

**Bands:**
Two degenerate 5f bands
One conduction band
(nearest neighbour tight-binding)
Hybridization $V_{fd}$ between the conduction and the 5f $\alpha$ band

$5f \alpha$ Characters of upper and lower hybridized bands
The 5f $\beta$ band is unhybridized
Note that for most $k$ values the $\alpha$ and $\beta$ bands have their relative energies shifted by an amount $V_{fd}^2$
(Depending on $\mu$, they have roughly the same nesting vectors, $Q$)
**Normal State Density of States**

Hybridized 5f–α states (blue)

Total hybridized f α plus conduction (d) band DOS (black)

Direct Hybridization Gap $\sim 2\sqrt{N} V_{fd}$ (below Fermi energy)

Unhybridized 5f-β states (red) (N-1)-fold degenerate

Chemical potential $\mu$ in the upper heavy α 5f band

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**Model for Hidden Order**

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Spin Rotationally Invariant Coulomb Interactions

The Coulomb interaction can be re-written in the form

\[ \hat{H}_{\text{int}} = \left( \frac{U - J}{2N} \right) \sum_{k, k', q, \sigma, \chi \neq \chi'} f_{k+q, \sigma}^{\dagger} f_{k, \sigma} f_{k', -q, -\sigma}^{\dagger} f_{k', -\sigma} \]

\[ + \left( \frac{U}{2N} \right) \sum_{k, k', q, \sigma, \chi, \chi'} f_{k+q, \sigma}^{\dagger} f_{k, \sigma} f_{k', -q, -\sigma}^{\dagger} f_{k', -\sigma} \]

\[ + \left( \frac{J}{2N} \right) \sum_{k, k', q, \sigma, \chi \neq \chi'} f_{k+q, \sigma}^{\dagger} f_{k, \sigma}^{\dagger} f_{k', -q, -\sigma} f_{k', -\sigma} \] (7)

U is the direct Coulomb Interaction

J is the Hund’s rule exchange

- \( J S_{\chi,i} \cdot S_{\chi',i} \)

As can be seen by commuting the annihilation operators, the last term is equivalent to the spin-flip part of the Hund’s rule exchange between orbital \( \chi \) and \( \chi' \)

\[ - \frac{J}{2} \left( S_{\chi,i}^+ S_{\chi',i}^- + S_{\chi,i}^- S_{\chi',i}^+ \right) \]

But we view it as a spin conserving hopping process involving a spin-up electron and a spin-down electron
A possibility for the hidden order parameter is $Z_Q$

$$Z_Q = \frac{1}{N} \sum_{k,\sigma} \sigma \left< f^{+\beta}_{k+Q,\sigma} f^{\alpha}_{k,\sigma} \right>$$

which is driven by the spin-flip part of the Hund’s rule exchange $J$. It only connect states with different orbital indices ($\alpha, \beta$).

(no net spin)

(broken spin-rotational invariance, and produces an $x$-$y$ anisotropy)

It can be complex (broken gauge invariance)

See also: Tanmoy Das, Scientific Reports, (2012).
A Mean-Field Approximation

- Linearize the (spin-flip Hund’s rule) interactions (momentum indices suppressed)

\[ + J f^{+,\beta \uparrow} f^{\alpha \uparrow} < f^{+,\alpha \downarrow} f^{\beta \downarrow} >_Q + J f^{+,\alpha \downarrow} f^{\beta \downarrow} < f^{+,\beta \uparrow} f^{\alpha \uparrow} >_Q \]
\[ - J < f^{+,\beta \uparrow} f^{\alpha \uparrow} >_Q < f^{+,\alpha \downarrow} f^{\beta \downarrow} >_Q \]
\[ + \text{Hermitean conjugate} \]

What if? \[ < f^{+,\beta \uparrow} f^{\alpha \uparrow} >_Q = - < f^{+,\beta \downarrow} f^{\alpha \downarrow} >_Q \]

(1) The energy would be lowered by \( \Delta E \) compared to the normal state defined by

\[ < f^{+,\beta} f^{\alpha} >_Q = 0 , \quad \Delta E = - \sum_\sigma J \left| < f^{+,\beta}_\sigma f^{\alpha}_\sigma >_Q \right|^2 \]

(2) **Spin-dependent (inter-orbital) Hybridization:** (with momentum transfer \( Q \))

\[ H_{hyb} = + \sum_k ( J < f^{+,\beta \uparrow} f^{\alpha \uparrow} >_Q f^{+,\alpha \downarrow}_{k-Q \downarrow} f^{\beta \downarrow}_{k \downarrow} + \text{H.c.} ) \]
\[ - \sum_k ( J < f^{+,\beta \uparrow} f^{\alpha \uparrow} >_Q f^{+,\alpha \downarrow}_{k-Q \uparrow} f^{\beta \uparrow}_{k \uparrow} + \text{H.c.} ) \]
A Mean-Field Approximation

\[
a^+_k,\uparrow = \sqrt{\frac{1}{2}} \left( f^{+,\alpha}_{k+Q,\uparrow} + f^{+,\beta}_{k,\uparrow} \right) \\
a^+_k,\downarrow = \sqrt{\frac{1}{2}} \left( f^{+,\alpha}_{k+Q,\downarrow} - f^{+,\beta}_{k,\downarrow} \right)
\]

(spin-dependent hybridized 5f bands)
Not sensitive to an (spin-independent) orbital measurement

\[
\frac{1}{2} \mid \Psi^\alpha_{k+Q} + \Psi^\beta_k \mid^2 + \frac{1}{2} \mid \Psi^\alpha_{k+Q} - \Psi^\beta_k \mid^2
\]

\[
= \mid \Psi^\alpha_{k+Q} \mid^2 + \mid \Psi^\beta_k \mid^2
\]

(same result as in the normal state where there are no interference terms)
The spin-up orbital density wave is compensated by a spin down orbital density wave.

 Requires Fermi-surface nesting in the normal state!
The states on the Fermi-surfaces of the $\alpha$ and $\beta$ bands are connected by the nesting vector $Q$

Interband:
$\beta$–$\alpha$ red to blue

Intraband:
$\alpha$–$\alpha$ blue to blue
$\beta$–$\beta$ red to red

Note: The red $\beta$ and blue $\alpha$ 5f bands are not degenerate but are shifted by a hybridization gap with a very small energy of the order $V_{fd}^2/W$.

Fermi-surface Interband Nesting

• Effect of energy shift due to hybridization on inter and intraband nesting: relative shift of Fermi-energy

Schematic in 2d:
The red $\beta$ and blue $\alpha$ 5f bands are not degenerate but are shifted by a very small energy of the order $V_{fd}^2/W$. 
The Linearized Gap Equation

\[\begin{align*}
1 - (U - J)\chi_{f,\sigma}^{\alpha,\beta,(0)}(Q, 0) & \quad z_{Q,\sigma}^* = -z_{Q,-\sigma}^* J \chi_{f,\sigma}^{\alpha,\beta}(Q, 0), \\
1 - (U - J)\chi_{f,-\sigma}^{\alpha,\beta,(0)}(Q, 0) & \quad z_{Q,-\sigma}^* = -z_{Q,\sigma}^* J \chi_{f,-\sigma}^{\alpha,\beta}(Q, 0),
\end{align*}\]

where

\[\chi_{f,\sigma}^{\alpha,\beta,(0)}(Q, 0) = \frac{1}{N} \sum_{k,\pm} |A_{\sigma}^{\pm}(k)|^2 \left( f\left[E_{\sigma}(k)\right] - f\left[E_{f,\sigma}(k + Q)\right] \right).\]

1. The equations are odd in \(z\) and possess a trivial solution \(z_{Q,\sigma} = 0\) for \(T > T_{HO}\).
2. The interband susceptibility \(\chi^{\alpha_\beta}(Q, 0)\) is positive, and \textbf{large}, if there is \textbf{inter-band nesting}.
3. The \textbf{Hund’s rule J exchange} is \textbf{enhanced} by the Coulomb interaction \(U\),
   \[1 = J \chi^{\alpha_\beta}(Q)/[1-(U-J) \chi^{\alpha_\beta}(Q)]\]
4. At the \textbf{critical temperature} \(T = T_{HO}\), one has an (infinitesimal) non-zero solution with \(z_{Q,\sigma} = -z_{Q,-\sigma}\).
Nesting and Adiabatic Continuity

Fig. 3. (Color online) Band characters of the energy dispersions of URu$_2$Si$_2$ in the $bc$ BZ. Green/light gray symbols illustrate the $\delta f_{\pi/2}$ $j_z = \pm 3/2$ character, blue/dark gray symbols the $j_z = \pm 3/2$ character, and red/gray symbols the $j_z = \pm 1/2$ character (for the symmetry points used, see Ref. 35).

LDA Peter Oppeneer (2011)

Two nesting vectors in URu$_2$Si$_2$: One is Commensurate and one is Incommensurate.

dHvA Hassinger (2010)

$Q_0=(0,0,1)$

Hidden Ordering, Magnetic Ordering

$Q_0=(0,0,1)$
Adiabatic Continuity?

- Change $\mu$ (fixed $V_{fd}$)
  Criterion for Instability ($U=J$)
  $1/U = \chi^{\alpha\alpha}(Q,0) + \chi^{\beta\beta}(Q,0)$
  **Antiferromagnetism**
  **Hidden Order**
  $1/U = \chi^{\alpha\beta}(Q,0)$
  **Antiferromagnetism**
  (separated by $\mu$ the order of $V_{fd}^2/W$)

- Adiabatic Continuity:
  either $V_{fd} \rightarrow 0$ or $W$ increases
  AF and HO instabilities become degenerate
The Gap Equation

\[ \chi_{Q,\sigma}^+ = -\frac{1}{N} \sum_k \left[ \int_C \frac{d\omega}{2\pi i} f(\omega) G_{f_f,\sigma}^{\beta,\alpha}(k + Q, k, \omega) \right] \]

The Hartree-Fock band dispersion relation \( E_{f,\sigma}^X(k) \) is given by

\[ E_{f,\sigma}^X(k) = E_f^X(k) + \sum_{\chi'} \left( (U - J) n_{f,\sigma} \chi' (1 - \delta_{\chi,\chi'}) + U n_{f,\sigma}^\chi \right) \]

and the gap parameter \( \kappa_{Q,\sigma} \) is defined as the complex number

\[ \kappa_{Q,\sigma} = J z_{-Q,-\sigma} - (U - J) z_{Q,\sigma} \]

The mixed character \( 5f \) Green’s function is given by

\[ G_{f_f,\sigma}^{\beta,\alpha}(k, k', \omega) = \frac{\kappa_{Q,\sigma}^* (\omega - \epsilon(k + Q)) \delta_{\alpha,\chi'} \delta_{k + Q, k'}}{D_{\sigma}(k + Q, \omega)} \]

where the denominator is given by

\[ D_{\sigma}(k, \omega) = \left[ \left( \omega - E_{f,\sigma}^\beta(k + Q) \right) \left( \omega - E_{f,\sigma}^\alpha(k) \right) - |\kappa_{Q,\sigma}|^2 \right] \left( \omega - \epsilon(k) \right) \]

\[ - |V_{\alpha}(k)|^2 \left( \omega - E_{f,\sigma}^\beta(k + Q) \right) \]

\[ 2|\kappa(0)|/k_B T_c = 4.54 \]
f- Quasiparticle Bands

Fermi Energy $\mu$

Hidden Order Gaps
Asymmetric HO Gap in DOS

Anayajian et al. (2010)

60% Fermi Surface Gapped

Asymmetric HO Gap

The Hidden Order Transition produces a pseudo-gap in the DOS.
Magnetic Nematicity

- **Broken Spin Rotational Invariance** ($V=0$)
  
  Upper gap edge state (unoccupied)
  \[
  \left( \psi_{k+Q}^{\alpha} + \psi_{k}^{\beta} \right)/\sqrt{2} \quad \left( \psi_{k+Q}^{\alpha} - \psi_{k}^{\beta} \right)/\sqrt{2}
  \]

  Lower gap edge state (occupied)
  \[
  \left( \psi_{k+Q}^{\alpha} - \psi_{k}^{\beta} \right)/\sqrt{2} \quad \left( \psi_{k+Q}^{\alpha} + \psi_{k}^{\beta} \right)/\sqrt{2}
  \]

  Band Gap
  \[2J |z|\]

- **Zeeman Interaction** (orientational dependence wrt to the $z$ axis)

  **Parallel**
  \[- \mu_B H^z \sigma^z\]
  Matrix elements between occupied and unoccupied states are zero
  
  No field dependence of the Energy \( \therefore \chi = 0 \)

  **Perpendicular**
  \[- \mu_B H^x \sigma^x\]
  Matrix elements between occupied and unoccupied states are unity
  
  Field dependence of the Energy
  \[- \left( \mu_B H^x \right)^2 / 2J |z|\]
Magnetic Nematicity

• Perpendicular susceptibility $\Gamma$

Matrix elements $\mu_B J z / \sqrt{(\varepsilon(k)^2 + J^2 Z^2)}$

Gap $2 \sqrt{(\varepsilon(k)^2 + J^2 Z^2)}$

Susceptibility

$$\mu_B^2 \int d\varepsilon \rho(\mu) \frac{J^2 Z^2}{(\varepsilon^2 + J^2 Z^2)^{3/2}}$$

Nominally proportional to order parameter squared, but

$$\chi \sim 4 \mu_B^2 \rho(\mu)$$
Magnetic Nematicity

- Quasiparticle Dispersion Relations in Field (V=0)
  - Field Parallel to $z$: $2\mu_B H_z$
    Spin Split Bands
  - Field Perpendicular to $z$: Coupled Bands
Quantum Critical Point?

Continuous Transition ends with a line of First-order Transitions?

Marcelo Jaime et al. (2002).

On decreasing J one expects to reach a Quantum Critical Point. However:

Self consistency conditions for the gap as a function of $U^{-1}$ for different $T$, for $\mu$ slightly off from the ideal nesting value

$$U^{-1} = \chi^{\alpha\beta}(T,z_Q)$$

$U^{-1} < 14$ Second-order
$U^{-1} > 14$ First-order

$\mu = 0.318$

Riseborough&Magalhaes
QCP & Discontinuous Transition

Figure 4: The gap $\kappa_{Q,\sigma}$ as function of $k_B T$ for $J = 0.075$, $D = 0.60$ and different intensities of the magnetic field $h$.

Figure 5: Phase diagram as function of the magnetic field $h$.

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Conclusions

• For systems with more than one occupied 5f band, there may be order parameters corresponding to the spontaneous (spatially inhomogeneous) mixing of the 5f bands, i.e.

\[ \Sigma_{k,\sigma} \sigma \langle f_{k+Q,\sigma}^{+\beta} f_{k,\sigma}^{\alpha} \rangle \neq 0 \]

• The transition has broken spin-rotational invariance but doesn’t have a staggered moment.
(Magnetic Nematicity)

• The Hund’s rule exchange J may stabilize an inter-orbital spin density wave

\[ \Sigma_{k,\sigma} \sigma \langle f_{k+Q,\sigma}^{+\beta} f_{k,\sigma}^{\alpha} \rangle \neq 0 \]

• The pseudogap in the DOS has a magnitude of \( U z_{Q,\sigma} \)

• The Hund’ rule mechanism could equally apply to transition metals.
(eg Fe-pnictides, especially where there is magnetic nematicity.)
f Quasiparticle Bands

Weights and Dispersion

5f bands of $\alpha$ character

5f bands of $\beta$ character
Quasiparticle Conduction Bands

Bands and weights with d character