Spin freezing and unconventional superconductivity

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Generic phase diagram of unconventional superconductors
- Superconducting dome next to a magnetically ordered phase
- Non-Fermi liquid metal above the superconducting dome

\[ \Sigma(\omega) \sim \sqrt{\omega} \]
**Method**

- **Dynamical mean field theory DMFT**: mapping to an impurity problem

  ![Lattice and impurity models](image)

  \[ G_{\text{latt}} \equiv G_{\text{imp}} \]
  \[ \Sigma_{\text{latt}} \equiv \Sigma_{\text{imp}} \]

- **Impurity solver**: computes the Green's function of the correlated site

- **Bath parameters = “mean field”**: optimized in such a way that the bath mimics the lattice environment

*Georges and Kotliar, PRB (1992)*
CT-QMC solvers allow efficient simulation of multiorbital models

\[
H_{\text{loc}} = - \sum_{\alpha,\sigma} \mu n_{\alpha,\sigma} + \sum_{\alpha} U n_{\alpha,\uparrow} n_{\alpha,\downarrow} + \sum_{\alpha > \beta, \sigma} U' n_{\alpha,\sigma} n_{\beta,-\sigma} + (U' - J) n_{\alpha,\sigma} n_{\beta,\sigma} - \sum_{\alpha \neq \beta} J (\psi_{\alpha,\downarrow}^\dagger \psi_{\beta,\uparrow}^\dagger \psi_{\beta,\downarrow} \psi_{\alpha,\uparrow} + \psi_{\beta,\uparrow}^\dagger \psi_{\beta,\downarrow}^\dagger \psi_{\alpha,\uparrow} \psi_{\alpha,\downarrow} + \text{h.c.})
\]

Relevant cases:

- 4 electrons in 3 orbitals: \textit{Sr}_2\textit{RuO}_4
- 3 electrons in 3 orbitals, \(J<0\): \textit{A}_3\textit{C}_{60}
- 6 electrons in 5 orbitals: \textit{Fe}-pnictides
Phase diagram for $U' = U - 2J, J/U = 1/6, \beta = 50$

Metallic phase: “transition” from Fermi liquid to incoherent metal

Narrow crossover regime with self-energy

$\text{Im} \Sigma/t \sim (i\omega_n/t)^\alpha, \alpha \approx 0.5$
Fit self-energy by $-\text{Im}\Sigma(i\omega_n) = C + A(\omega_n)^{\alpha}$

Square-root self-energy coincides with on-set of frozen moments
Strontium Ruthenates

A self-energy with frequency dependence $\Sigma(\omega) \sim \omega^{1/2}$ implies an optical conductivity $\sigma(\omega) \sim 1/\omega^{1/2}$

Non-Fermi-Liquid Behavior of SrRuO$_3$: Evidence from Infrared Conductivity

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(Received 13 March 1998)

The reflectivity of the itinerant ferromagnet SrRuO$_3$ has been measured between 50 and 25 000 cm$^{-1}$ at temperatures ranging from 40 to 300 K, and used to obtain conductivity, scattering rate, and effective mass as a function of frequency and temperature. We find that at low temperatures the conductivity falls unusually slowly as a function of frequency (proportional to $1/\omega^{1/2}$), and at high temperatures it even appears to increase as a function of frequency in the far-infrared limit. The data suggest that the charge dynamics of SrRuO$_3$ are substantially different from those of Fermi-liquid metals.
Spin-freezing

Spin-spin and orbital-orbital correlation functions

-0.1
-0.05
0
0.05
0.1
0.15
0.2
0.25

\( \langle n_1(0)n_2^{z}(\tau) \rangle \), \( \langle S_z(0)S_z(\tau) \rangle \)

\( n=1.21 \)
\( n=1.75 \)
\( n=2.23 \)
\( n=2.62 \)
\( n=2.97 \)

Freezing of spin moments

No freezing of orbital moments

Werner, Gull, Troyer & Millis
PRL 101, 166405 (2008)
Consider the local susceptibility

\[ \chi_{\text{loc}} = \int_0^\beta d\tau \langle S_z(\tau)S_z(0) \rangle \]

and its dynamic contribution

\[ \Delta \chi_{\text{loc}} = \int_0^\beta d\tau [\langle S_z(\tau)S_z(0) \rangle - \langle S_z(\beta/2)S_z(0) \rangle ] \]

subtract the (frozen) long-time value
Consider the local susceptibility $\chi_{loc}$ and its dynamic contribution $\Delta \chi_{loc}$.

Crossover regime is characterized by large local moment fluctuations.

Crossover regime is characterized by large local moment fluctuations.
Pnictides

- Strongly correlated despite moderate $U$

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incoherent metal state resulting from Hund’s coupling

Haule & Kotliar, NJP (2009)
Strong doping and temperature dependence of electronic structure

\[ \text{BaFe}_2\text{As}_2: \]

- Conventional FL metal in the underdoped regime
- Non-FL properties near optimal doping
- Incoherent metal in the overdoped regime
Identify ordering instabilities by divergent lattice susceptibilities

- Calculate local vertex from impurity problem
- Approximate vertex of the lattice problem by this local vertex
- Solve Bethe-Salpeter equation to obtain lattice susceptibility

The following orders (staggered and uniform) are considered:

- **diagonal orders:**
  charge, spin, orbital, spin-orbital

- **off-diagonal orders:**
  orbital-singlet-spin-triplet SC, orbital-triplet-spin-singlet SC

Hoshino & Werner
PRL 115, 247001 (2015)
- 3-orbital model, Ising interactions

AFM near half-filling
FM at large U away from half-filling
spin-triplet superconductivity in the spin-freezing crossover region
3-orbital model, Ising interactions (lower temperature)

AFM near half-filling

FM at large U away from half-filling

spin-triplet superconductivity in the spin-freezing crossover region

parameter regime relevant for $\text{Sr}_2\text{RuO}_4$
$T_c$ dome and non-FL metal phase next to magnetic order

Generic phasediagram of unconventional SC without QCP!
$T_c$ dome and non-FL metal phase next to magnetic order

Need spin-anisotropy (SO coupling) for high $T_c$
probably relevant for: Sr$_2$RuO$_4$, UGe$_2$, URhGe, UCoGe, ...
**Long-range order**

**Hoshino & Werner**

*PRL 115, 247001 (2015)*

- **Pairing induced by local spin fluctuations**
  
  Weak-coupling argument inspired by Inaba & Suga, PRL (2012)

- **Effective interaction which includes bubble diagrams:**
  
  \[ \tilde{U}_{\alpha\beta}(q) = U_{\alpha\beta} - \sum_{\gamma} U_{\alpha\gamma} \chi_{\gamma}(q) \tilde{U}_{\gamma\beta}(q) \]

  ![Effective interaction diagram]

- **Effective inter-orbital same-spin interaction**
  
  \[ \tilde{U}_{1\uparrow,2\uparrow}(0) = U' - J - [2UU' + (U' - J)^2 + U''^2] \chi_{\text{loc}} \]

  *in the weak-coupling regime: \( \chi_{\text{loc}} = \Delta \chi_{\text{loc}} \)*
Negative J and orbital freezing

- 2-orbital model ($U=\text{bandwidth}=4$)

Steiner et al. PRB 94, 075107 (2016)
Away from half-filling: SC dome peaks near orbital freezing line
**Negative $J$ and orbital freezing**

- **Half-filled 3-orbital model with $J<0$ ($A_3C_{60}$)**

  - Fermi liquid metal
  - Orbital frozen metal
  - Mott insulator

  - SC dome peaks in the region of maximum orbital fluctuations
  - Spontaneous symmetry breaking into an orbital selective Mott phase ("Jahn-Teller metal")

Hoshino & Werner
PRL 118, 177002 (2017)
Mapping to an effective two-orbital model:

\[ c_1 = \frac{1}{\sqrt{2}} (d_1 + d_3) \quad c_2 = \frac{1}{\sqrt{2}} (d_2 + d_4) \]
\[ f_1 = \frac{1}{\sqrt{2}} (d_1 - d_3) \quad f_2 = \frac{1}{\sqrt{2}} (d_2 - d_4) \]

Slater-Kanamori interaction with \( \tilde{U} = \tilde{U}' = \tilde{J} = U/2 \)

nnn hopping translates into a crystal-field splitting \( \delta = 2t' \)
Mapping to an effective two-orbital model:

\[ c_1 = \frac{1}{\sqrt{2}} (d_1 + d_3) \quad c_2 = \frac{1}{\sqrt{2}} (d_2 + d_4) \]
\[ f_1 = \frac{1}{\sqrt{2}} (d_1 - d_3) \quad f_2 = \frac{1}{\sqrt{2}} (d_2 - d_4) \]

Slater-Kanamori interaction with \( \tilde{U} = \tilde{U}' = \tilde{J} = U/2 \)
nnn hopping translates into a crystal-field splitting \( \delta = 2t' \)
Phasediagram (2-site/2-orbital cluster DMFT)

- emerging (fluctuating)
- local moments = bad metal regime

- frozen moments = pseudo-gap phase

Cuprates

Hoshino & Werner (2016)
Cuprates

- Phasediagram (2-site/2-orbital cluster DMFT)

- Emerging (fluctuating) local moments = bad metal regime

- SC dome [4-site cluster DMFT, Maier et al, (2005)] induced by fluctuating local moments?
Cuprates

- d-wave SC induced by local spin fluctuations

- Transformation of the d-wave order parameter:

\[
(d_{1\uparrow}^\dagger d_{2\downarrow}^\dagger - d_{1\downarrow}^\dagger d_{2\uparrow}^\dagger) - (d_{2\uparrow}^\dagger d_{3\downarrow}^\dagger - d_{2\downarrow}^\dagger d_{3\uparrow}^\dagger) \\
+ (d_{3\uparrow}^\dagger d_{4\downarrow}^\dagger - d_{3\downarrow}^\dagger d_{4\uparrow}^\dagger) - (d_{4\uparrow}^\dagger d_{1\downarrow}^\dagger - d_{4\downarrow}^\dagger d_{1\uparrow}^\dagger) \rightarrow 2(f_{1\uparrow}^\dagger f_{2\downarrow}^\dagger - f_{1\downarrow}^\dagger f_{2\uparrow}^\dagger)
\]

- Effective attractive interaction:

\[
\tilde{U}_{\text{eff}}^{(1,f,\uparrow),(2,f,\downarrow)} = 2\tilde{U}^3 \chi_{\text{loc}}^{(f)} \chi_{12}^{(c)} + O(\tilde{U}^5)
\]

- Leading contribution:

\[
\begin{array}{c}
1f \uparrow \\
\bowtie
\end{array}
\quad =
\begin{array}{c}
1f \uparrow \\
\tilde{U}_{\text{loc}}^{(f)} \chi_{12}^{(c)}
\end{array}
\begin{array}{c}
2f \downarrow \\
\bowtie
\end{array}
\quad =
\begin{array}{c}
2f \downarrow \\
\tilde{U}_{\text{loc}}^{(f)} \chi_{12}^{(c)}
\end{array}
\begin{array}{c}
1c \uparrow \\
\tilde{U}'
\end{array}
\begin{array}{c}
2c \uparrow \\
\tilde{U}'
\end{array}
\begin{array}{c}
2f \downarrow \\
\tilde{U}'
\end{array}
\end{array}
\]
Further evidence

- Spin correlations in the 2D Hubbard model (DCA results)
- Compare nearest neighbor correlations ($S_{12}$) to diagonal next-nearest neighbor correlations ($S_{13}$)

![Graphs showing spin correlations in 4-site and 8-site DCA calculations](image)

**Note:** The graphs illustrate the decay of spin correlations ($S_{12}$ and $S_{13}$) with increasing imaginary time ($\tau$) for different fillings ($n=0.5, 0.42, 0.36$). The red line represents $n=0.5$, the blue line $n=0.42$, and the black line $n=0.36$. The 4-site and 8-site DCA results are compared, with the former showing a faster decay for $S_{12}$ compared to $S_{13}$, indicating a stronger antiferromagnetic nearest-neighbor interaction. At long imaginary times, ferromagnetic correlations dominate, as highlighted by the red arrow on the graph.
Further evidence

- **Spin correlations in the 2D Hubbard model (DCA results)**

- **Plot** $\beta[S_{13} - (-S_{12})]$ as a function of temperature and filling

![Diagram showing the dependence of $\beta[S_{13} - (-S_{12})]$ on temperature ($T$) and filling per spin.](image)

**4-site**

- Robust FM correlations (formation of composite spin-1)
Spin correlations in the 2D Hubbard model (DCA results)

Plot $\beta [S_{13} - (-S_{12})]$ as a function of temperature and filling

Appearance of composite spin-1 as origin of the pseudo-gap
Summary

- Spin/orbital freezing as a universal phenomenon in unconventional superconductors
  - Strontium ruthenates
  - Uranium-based SC
  - Pnictides
  - Fulleride compounds
  - Cuprates
  - ...

- Pairing induced by local spin or orbital fluctuations

- Bad metal physics originates from fluctuating/frozen moments
3-orbital model

“quasi-particle weight” $z$

from De’ Medici, Mravlje & Georges, PRL (2011)

large local moment fluctuations

Hund coupling $J$: Strongly correlated metal far from the Mott transition
Strong doping and temperature dependence of electronic structure
2-orbital model \((U=\text{bandwidth}=4)\)

- Mapping between \(J<0\) and \(J>0\):

\[
\begin{pmatrix}
  d_{i,1\downarrow} \\
  d_{i,2\uparrow}
\end{pmatrix}
\longrightarrow
\begin{pmatrix}
  0 & 1 \\
  1 & 0
\end{pmatrix}
\begin{pmatrix}
  d_{i,1\downarrow} \\
  d_{i,2\uparrow}
\end{pmatrix}
\]

\(J<0\):
- spin-singlet SC
- antiferro OO
- ferro OO
- orbital freezing

\(J>0\):
- spin-triplet SC
- AFM
- FM
- spin freezing
Phasediagram (1-site/2-orbital DMFT)

- Emerging (fluctuating) local moments = bad metal regime
- Frozen moments = pseudo-gap phase

Cuprates

Werner, Hoshino & Shinaoka
PRB 94, 245134 (2016)
Further evidence

- Spin correlations in the 2D Hubbard model (DCA results)
- Plot $\beta [S_{13} - (-S_{12})]$ as a function of temperature and filling
- Appearance of composite spin-1 as origin of the pseudo-gap

![Diagram showing fluctuation of spin correlations](image-url)
Further evidence

- Spin correlations in the 2D Hubbard model (DCA results)
- Plot $\beta[S_{13} - (-S_{12})]$ as a function of temperature and filling

![Graph showing spin correlations](Image)

- Appearance of composite spin-1 as origin of the pseudo-gap