Progress on Itinerant Electrons – Ferromagnetism, Curie-Weiss Metal, and Spin-orbit Ordering

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Thank J. Hirsch, I. Schuller, S. Kivelson, Lu Yu, Tin-Lun Ho for helpful discussions.

2) S. L. Xu, Yi Li, C. Wu, PRX 5, 021032 (2015).
3) Yi Li, C. Wu, PRB 85, 205126 (2012).
The early age of ferromagnetism

The magnetic stone attracts iron.

慈 (ci) 石(shi) 召(zhao) 铁(tie)

---- Guiguzi (鬼谷子), (4th century BC)

(loving, merciful, compassionate):
the original Chinese character for magnetism

heart

磁
magnetism, magnetic
stone

Thales says that a stone (lodestone) has a soul because it causes movement to iron.

---- De Anima, Aristotle (384-322 BC)

“Slightly eastward, not directly south” (常微偏东,不全南也)- Kuo Shen (沈括)(1031-1095)

World’s first compass:
magnetic spoon: 1 century AD (司南 South-pointer)
Magnetism of Itinerant Electrons

- Quantum Origin
- Strong correlation
- Ferromagnetism, Hund’s rule, Curie-Weiss metal
- Unconventional symm.
- p-wave magnetism spin-orbit, multipolar ordering
Local moments v.s. itinerant electrons

- Local moments – **not our interest!**

\[ H = -J \sum_{ij} \sigma_i \sigma_j \]

Curie-Weiss susceptibility

\[ \chi = \frac{A}{T - T_c} \]

- Itinerant electrons: Fermi surfaces – much harder to form ferromagnetism!

Pauli paramagnetism

\[ \chi = N_0 \left(1 - \frac{T^2}{T_f^2}\right) \]

- \(N_0\): density of states at the Fermi level
- \(N_0\): density of states at Fermi energy
Itinerant ferromagnetism – Fermi statistics

- Exchange due to exclusion – polarized electrons avoid each other.

- Stoner criterion: kinetic energy cost.

\[ E_{\uparrow\uparrow} < E_{\uparrow\downarrow} \]

\[ UN_0 > 1 \]

\[ U \] – average interaction strength; \( N_0 \) – density of states at the Fermi level
Kohn-Sham density functional theory – Work at Univ. California, San Diego

- Accurate on the ground state magnetizations.

<table>
<thead>
<tr>
<th>Property</th>
<th>source</th>
<th>Fe (bcc)</th>
<th>Co (fcc)</th>
<th>Ni (fcc)</th>
<th>Gd (hep)</th>
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<tr>
<td>$M_{\text{spin}}$</td>
<td>LSDA</td>
<td>2.15</td>
<td>1.56</td>
<td>0.59</td>
<td>7.63</td>
</tr>
<tr>
<td>$M_{\text{spin}}$</td>
<td>GGA</td>
<td>2.22</td>
<td>1.62</td>
<td>0.62</td>
<td>7.65</td>
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<td>$M_{\text{spin}}$</td>
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<td>2.12</td>
<td>1.57</td>
<td>0.55</td>
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<td>experiment</td>
<td>2.22</td>
<td>1.71</td>
<td>0.61</td>
<td>7.63</td>
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</table>

- Correlations partially contained in $V_{xc}(r)$ for energetics.

But wavefunctions remain Slater-determinant (uncorrelated) type.

- Thermal fluctuations difficult to handle – Curie temperatures overestimated.
Itinerant ferromagnetism v.s. superconductivity

<table>
<thead>
<tr>
<th>I</th>
<th>H</th>
<th>Li</th>
<th>Be</th>
<th>Na</th>
<th>Mg</th>
<th>K</th>
<th>Ca</th>
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<tr>
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**KNOWN SUPERCONDUCTIVE ELEMENTS**

- **BLUE = AT AMBIENT PRESSURE**
- **GREEN = ONLY UNDER HIGH PRESSURE**

**Lanthanide Series**

<table>
<thead>
<tr>
<th>58</th>
<th>Ce</th>
<th>59</th>
<th>Pr</th>
<th>60</th>
<th>Nd</th>
<th>61</th>
<th>Pm</th>
<th>62</th>
<th>Sm</th>
<th>63</th>
<th>Eu</th>
<th>64</th>
<th>Gd</th>
<th>65</th>
<th>Tb</th>
<th>66</th>
<th>Dy</th>
<th>67</th>
<th>Ho</th>
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**Actinide Series**

<table>
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<tr>
<th>90</th>
<th>Th</th>
<th>91</th>
<th>Pa</th>
<th>92</th>
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<th>Cm</th>
<th>97</th>
<th>Bk</th>
<th>98</th>
<th>Cf</th>
<th>99</th>
<th>Es</th>
</tr>
</thead>
</table>

**FM elements:**

Fe, Co, Ni, Gd, Dy
Outline

Quantum Origin

Magnetism of Itinerant Electrons

Strong correlation

Ferromagnetism, Curie-Weiss metal

Unconventional symm.

New states: p-wave magnetism (spin-orbit ordering)
Correlations – FM is still rare!

- Electrons with opposite spins still avoid each other → Kinetic energy advantage via correlated wavefunctions.

- No go! Correlation wins: (I) two electrons, (II) 1D.

**Singlet (the ground state)**

\[ \phi_{\text{sym}}(x_1 - x_2) \]

(correlated, nodeless)

**Triplet**

\[ \phi_{\text{asym}}(x_1 - x_2) \]

(exchange, nodal)

No FM in 1D – Lieb & Mattis theorem: spin singlet ground states.
Local v.s. global: Hund’s coupling ≠ ferromagnetism

- Most ferro-metals: orbital degeneracy, Hund’s coupling.
- Electron/hole spins add up – the exchange interaction.

- Hund’s rule usually cannot polarize the entire lattice!

- Under what condition, Hund’s rule → global ferromagnetic coherence?
Curie-Weiss metal $\leftrightarrow$ high $T_c$ pseudo-gap phase

- Curie-Weiss susceptibility: $\chi = \frac{A}{1 - T/T_0}$, $T_0 < T \ll T_f$

  Natural for local moments but not for Fermi surfaces!

- Paramagnetic states close to $T_0$: domain fluctuations!

  $T > T_0$

  $\neq$

  Stronger correlation than in the polarized FM state
Non-perturbative study on itinerant FM

A simple and quasi-realistic model

An entire phase of ground state FM

Controllable study of Curie-Weiss metal

Quantum Monte-Carlo (sign-problem free)

Mechanism and experiments

Theorem proof


S. Xu, Yi Li, and C. Wu, PRX 5, 021032, (2015).
Hund’s rule + quasi-1D bands (p/d-orbitals) $\rightarrow$ 2D and 3D FM in the strong interaction regime.

- $d_{xz}/d_{yz}$ in 2D transition metal oxides
- $p_x$, $p_y$, ($p_z$) in 2D or 3D optical lattices.
Multi-orbital onsite (Hubbard) interactions

- Intra-orbital repulsion $U \rightarrow \infty$.
  - Intra-orbital singlet projected out

- Inter-orbital **Hund’s coupling** $J>0$, and repulsion $V$.

\[
E = J + V
\]

\[
E = V
\]
Theorem for a **phase** of itinerant FM

- **Theorem**: FM ground states at $U \to \infty$ (fully polarized and unique up to $2S_{\text{tot}}+1$-fold spin degeneracy).

- **An entire phase**: valid at any generic filling, any value for $J>0$, and $V$.

- Free of quantum Monte-Carlo (QMC) sign problem at any filling – a rare case for fermions.

**A reliable reference point for analytic and numeric studies of FM in multi-orbital systems**

Hund’s rule assisted global FM

• Intra-chain:
  Finite $U$: singlet ground state
  $U \rightarrow \infty$: infinite degeneracy

• Inter-chain:
  Hund’s $J$ lifts the degeneracy
  $\rightarrow$ global FM.

• 2D FM coherence: Total spin in each chain not conserved.

\[
\begin{align*}
E & \quad S = N/2 \\
\vdots & \\
S & = 1 \\
S & = 0
\end{align*}
\]
Quantum Monte Carlo (QMC)

- Stochastic method – polling the Hilbert space with importance sampling.

\[ Z = Tr(e^{-\beta H}) \]

\[ = \sum \left\{ \langle \psi, \tau | (-H)^{g(b,\tau)} | \psi, \tau + d\tau \rangle \right\} \]

- Quantum Monte Carlo (QMC)

- Always Positive!

- Stochastic series expansion (SSE) + direct loop update.

- Our model is free of the sign problem – a rare case of fermion models.

• Local moment-like spin susceptibility (spin incoherent):

\[ \chi = \frac{A}{1 - T/T_0}, \quad T_0 < T < T_{ch} \]

• Metallic compressibility \( \kappa \):
Saturates at \( T < T_{ch} \), and \( T_{ch} \sim t \) (charge itinerancy).
Curie temperatures v.s filling (V=0)

\[ \chi = \frac{A}{1 - T/T_0} \]

- \( T_0 \to 0 \) at both \( n \to 0 \) (particle vaccum), and \( n \to 2 \) (hole vaccum, spin-1 moments, no FM).

- \( T_{0,max} \approx 0.08t_\parallel \) at maximal itinerancy (p-h symmetry)

\[ V = 0, J = 2 \]
Critical ferromagnetic scaling

- No long-range order at finite $T$ (Mermin-Wagner theorem)

- $O(3)$ NL$\sigma$-model: FM directional fluctuations

- As $T < T_0$, $\chi$ crosses over into an exponential growth.

\[
\chi = \frac{C}{T - T_0} \rightarrow \chi = A e^{b \frac{T_0}{T}}
\]
Fermi distribution $n_F(k)$

Paramagnetic Curie-Weiss metal

$n_F(k) = n_{\uparrow}(k) + n_{\downarrow}(k)$

- At $k \to 0$, $n_{\uparrow}(k) = n_{\downarrow}(k) \approx 0.54 \ll 1$
- Large entropy (the k-space picture)
- Strongly correlated metal phase

Reference: polarized fermion with $k_F^0 = \frac{\pi}{2}$
Calculation of $T_c$ for the Ising class!

Structure factor scaling

- Reduce symm to the Ising class.
  \[ J_{\parallel} = 2, \quad J_{\perp} = 4 \]

- $T_c$ is be suppressed from based on CW-law by non-Gaussian fluctuations.

\[
T_c = \frac{1}{\beta_c} = \frac{1}{7.6} \approx 0.132, \\
T_0 \approx 0.2
\]
Critical scaling and data collapse: 2D Ising class

Data crossing: \( S_\perp(0)L^{-2+\eta} \) v.s. \( T \)

\[
S_\perp(0)L^{-2+\eta} = f\left((T / T_c - 1)L^{1/\nu}\right)
\]

\[
\eta = \frac{1}{4}, \quad \nu = 1
\]

\( T_c \approx 0.134 \),
Apply to the LaAlO$_3$/SrTiO$_3$ interface

- $d_{xz}, d_{yz}$: quasi-1D bands, higher energy.
- $d_{xy}$: quasi-2D band, lower energy.

- Our theorem to $d_{xz}, d_{yz}$ bands: quas-1D + Hund’s coupling + strong intra-orbital repulsion for 3d-orbitals.

- The FM $d_{xz}, d_{yz}$ -bands polarize the paramagnetic $d_{xy}$ - band.

- No local moments needed! Different from double exchange.

A similar proposal by Chen and Balents based on mean-field treatment of the coupling between $dxz/dyz$-bands.

Lu Li et al, Nat. Phys 2011, Bert et al, Nat. Phys 2011
FM at the interface of SrTiO$_3$/LaAlO$_3$

Lu Li et al, Nat. Phys 2011, Bert et al, Nat. Phys 2011
Why is ferromagnetism difficult?

• Large kinetic energy cost to polarize the ideal Fermi distribution.

• Hund J is the key, but by itself, it is insufficient!

• Hubbard U mostly favors anti-FM, but brutal enough to distort the Fermi distribution.

• Apply J on top of U $\rightarrow$ FM with less kinetic energy cost and even gain kinetic energy.

![Graph showing changes in Fermi distribution with and without J and U](image-url)
Outline

Quantum Origin

Magnetism of Itinerant Electrons

- New states: p-wave magnetism (spin-orbit ordering)
- Unconventional symm.

Strong correlation

Ferromagnetism, Curie-Weiss metal
Itinerant ferromagnetism: \textbf{s-wave}

- Spin rotational symmetry is broken.

- Orbital rotational symmetry is \textbf{NOT} broken: spin polarizes along \textbf{a fixed direction}.

\textit{cf.} Conventional superconductivity (s-wave)

Unconventional superconductivity (e.g. d-wave high $T_c$ cuprates)
New states of matter: unconventional magnetism!

- High partial-wave channel magnetism (e.g. $p$, $d$-wave...).

- Multi-polar spin distribution over the Fermi surface.

**anisotropic $p$-wave state**

**isotropic $p$-wave state**


Spin flips the sign as $\vec{k} \rightarrow -\vec{k}$.
Anisotropic unconventional magnetism: spin nemetic liquid phases!

- $p$-wave distortion of the Fermi surface.

- Spin dipole moment in momentum space (not in coordinate space).

$$\vec{n}_1 = \sum_{\vec{k}} \vec{s}_k \cos \theta_{\vec{k}} \neq 0$$

- Both orbital and spin rotational symmetries are broken.


The isotropic $\rho$-wave magnetic phase

- Helicity $\vec{\sigma} \cdot \vec{k}$ is a good quantum number.

- No net spin-moment; spin dipole moment in momentum space.

\[ \vec{n}_1 = \sum_k \vec{s}_k \cos \theta_k, \quad \vec{n}_2 = \sum_k \vec{s}_k \sin \theta_k \]

- Isotropic phase with SO coupling.

\[ H_{MF} = H_0 + \vec{n} \sum_k \psi^+ \vec{\sigma}_{\alpha \beta} \cdot \vec{k} \psi_\beta \]

\[ \vec{n} = |\vec{n}_1| = |\vec{n}_2| \]

C. Wu et al., PRL 93, 36403 (2004);
C. Wu et al., PRB PRB.75, 115103 (2007).
Dynamic generation of spin-orbit (SO) coupling!

- Conventional wisdom:
  - A single-body effect inherited from the Dirac equation

- New mechanism (many-body collective effect):
  - Generate SO coupling through unconventional magnetic phase transitions.

- Advantages: tunable SO coupling by varying temperatures; new types of SO coupling.
The subtle symmetry breaking pattern

- $\vec{J}$ is conserved, but $\vec{L}$, $\vec{S}$ are not separately conserved.
- Independent orbital and spin rotational symmetries.

- Relative spin-orbit symmetry breaking.

Leggett, Rev. Mod. Phys 47, 331 (1975)
Unconventional magnetism: Pomeranchuk instability in the spin channel

- An analogy to superfluid $^3$He-B (isotropic) and A (anisotropic) phases.
**cf. Superfluid $^3$He-B, A phases**

- $\rho$-wave triplet Cooper pairing.

\[ \Delta(k) = \Delta \hat{d}(k) = \Delta \hat{k} \]

- $^3$He-B (isotropic) phase.

A. J. Leggett, Rev. Mod. Phys 47, 331 (1975)
Pomeranchuk instability

- Fermi surface: elastic membrane.
- Stability:
  \[ \Delta E_K \propto (\delta n_i^{s,a})^2 \]
  \[ \Delta E_{\text{int}} \propto \frac{F_l^{s,a}}{2l + 1} (\delta n_i^{s,a})^2 \]
- Surface tension vanishes at:
  \[ F_l^{s,a} < -(2l + 1) \]
- Ferromagnetism: the \( F_0^a \) channel.
- Nematic electron liquid: the \( F_2^s \) channel.

I. Pomeranchuk

\[ \delta n_i \]
The $\beta$-phases: vortices in momentum space

- Perform global spin rotations, $A \rightarrow B \rightarrow C$.

L. Fu’s (PRL 2015): gyro ferroelectric muti-polar
Neutron spectra (The $\alpha$-phase)

- No *elastic* Bragg peaks.

- $\vec{n}_{1,2}$ couple with spin dynamically **at $T < T_c$** --
  coupling between Goldstone modes and spin-waves.

\[
L = (\vec{n}_1 \times \partial_t \vec{n}_1 + \vec{n}_2 \times \partial_t \vec{n}_2) \cdot \dot{\vec{S}}
\]
\[
\rightarrow \vec{n} \left( S_y \partial_t n_{1x} - S_x \partial_t n_{1y} \right)
\]

- *In-elastic*: resonance peaks develop **at $T<T_c$**.

\[
\text{Im } \chi_{s,\pm}(\vec{q}, \omega) \approx N_0 \omega_q^2 \delta(\omega^2 - \omega_q^2)
\]
A natural generalization of ferromagnetism

- The driving force is still exchange interactions, but in non-s-wave channels.

<table>
<thead>
<tr>
<th></th>
<th>s-wave</th>
<th>p-wave</th>
<th>d-wave</th>
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</thead>
<tbody>
<tr>
<td>SC/SF</td>
<td>Hg, 1911</td>
<td>³He, 1972</td>
<td>high T&lt;sub&gt;c&lt;/sub&gt;, 1986</td>
</tr>
<tr>
<td>magnetism</td>
<td>Fe, ancient time</td>
<td>?</td>
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- Optimistically, unconventional magnets may already exist.

  cf. Antiferromagnetic materials are actually very common in transition metal oxides. But they were not well-studied until neutron-scattering spectroscopy was available.
• Consistent with orbital ordering between dxz/dyz orbitals.

Parity breaking nematic phase in Cd$_2$ReO$_7$

- Signature of parity breaking through 2$^{\text{nd}}$ harmonic generation method.

Summary: Itinerant magnetism (ferro and beyond)

- Non-perturbative study on itinerant FM and Curie-Weiss metals

**Hund’s rule + quasi 1D + strong correlation**

**Sign-problem QMC simulations.**

- New states of matter: p-wave magnetism

Spontaneous generation of spin-orbit ordering.

\[ d_{xz} \quad d_{yz} \quad -t_{||} \]

\[ -t_{||} \]