Unconventional(nematic) Metamagnetism in the t_{2g}-orbital System of Sr₃Ru₂O₇

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Ref: W. C. Lee, C. Wu, PRB 80, 104438 (2009);

W. C. Lee, C. Wu, PRL 103, 176101 (2009);

W. C. Lee, D. Arovas, and C. Wu, PRB 81, 184403 (2010);

W. C. Lee, C. Wu, arXiv:1008.2486.

Related past work on unconventional magnetism with S. C. Zhang (Stanford), E. Fradkin (UIUC).

C. Wu, S. C. Zhang, PRL 93, 36403 (2004); C. Wu, K. Sun, E. Fradkin, and S. C. Zhang, Phys. Rev. B 75, 115103 (2007).

Acknowledgments: J. C. Davis, J. Hirsch, S. Kivelson, A. Mackenzie.

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Outline

• Introduction to unconventional magnetism (Pomeranchuk instabilities with spin):

anisotropic states: nematic electron liquids with spin; isotropic states: spontaneous generation of spin-orbit coupling.

C. Wu, S. C. Zhang, PRL 93, 36403 (2004); C. Wu, K. Sun, E. Fradkin, and S. C. Zhang, Phys. Rev. B 75, 115103 (2007).

• Experimental results: nematic meta-magnetism in the t_{2g} orbital system of Sr₃Ru₂O₇.

• Microscopic theory with quasi-1D bands of d_{xz} and d_{yz}. Orbital degree of freedom facilitates unconventional metamagnetism exhibiting orbital ordering.

• STM quasi-particle interference as a test of orbital ordering.

Ferromagnetism: many-body collective effect



• Driving force: **exchange interaction among electrons**.





• Stoner criterion:

 $UN_{0} > 1$

E. C. Stoner



U – average interaction strength; N_0 – density of states at the Fermi level



Ferromagnetism: *s*-wave magnetism

• Spin rotational symmetry is broken.

 Orbital rotational symmetry is NOT broken: spin polarizes along a fixed direction on the Fermi surface.

• cf. conventional superconductivity.

Cooper pairing between electrons with opposite momenta.

s-wave: pairing amplitude does not change over the Fermi surface.





cf. Unconventional superconductivity

• High partial wave channel Cooper pairings (e.g. p, d-wave ...).

• *d*-wave: high T_c cuprates. Paring amplitude changes sign in the Fermi surface.



• *p*-wave: Sr₂RuO₄, ³He-A and B.

D. J. Van Harlingen, Rev. Mod. Phys. 67, 515 (1995); C. C. Tsuei et al., Rev. Mod. Phys. 72, 969 (2000).

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New states of matter: unconventional magnetism!

- High partial wave channel generalizations of FM (e.g. *p*, *d*-*wave*...) as spin-dependent **Pomeranchuk** instabilities.
- Spin polarization varies over the Fermi surface.



anisotropic (S) *p*-wave magnetic state

isotropic (බා *p*-wave magnetic state

spin flips the sign as $\vec{k} \rightarrow -\vec{k}$.

Anisotropy: liquid crystalline order

• Classic liquid crystal.

Nematic phase: rotational anisotropic but translational invariant.



• Quantum version of liquid crystal: **nematic electron liquid**.



S. Kivelson, et al, Nature 393, 550 (1998); V. Oganesyan, et al., PRB 64,195109 (2001). 7

<u>Anisotropic unconventional (S) magnetism:</u> <u>electron liquid crystal phases with **spin**!</u>



• *p*-wave distortion of the Fermi surface.

• No net spin-moment: $\vec{S} = \sum_{\vec{i}} \vec{s}_k = 0$

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

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

anisotropic *p*-wave magnetic phase

spin-split state by J. E. Hirsch, PRB 41, 6820 (1990); PRB 41, 6828 (1990). • Both orbital and spin rotational symmetries are broken.

V. Oganesyan, et al., PRB 64,195109 (2001). C. Wu et al., PRL 93, 36403 (2004); Varma et al., Phys. Rev. Lett. 96, 036405 (2006)

The isotropic (Δ) *p*-wave magnetic phase



C. Wu et al., PRL 93, 36403 (2004); C. Wu et al., PRB 75, 115103(2007).

- Spin is not conserved; helicity $\vec{\sigma} \cdot \vec{k}$ is a good quantum number.
- No net spin-moment; spin dipole moment in momentum space.

$$\vec{n}_1 = \sum_{\vec{k}} \vec{s}_k \cos \theta_k, \quad \vec{n}_2 = \sum_{\vec{k}} \vec{s}_k \sin \theta_k$$

• Relative spin-orbit symmetry breaking. Spontaneous generation of spin-orbit coupling without relativity!

$$H_{MF} = H_0 + \overline{n} \sum_{k} \psi_{\alpha}^{+} \vec{\sigma}_{\alpha\beta} \cdot \vec{k} \psi_{\beta}$$
$$\overline{n} = |\vec{n}_1| = |\vec{n}_2|$$

cf. p-wave pairing Superfluid ³He-B, A phases



• ³He-B (isotropic) phase.

• ³He-A (anisotropic) phase.

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

<u>2D</u> *d*-wave α and β -phases



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• Experiments: nematic meta-magnetism in the t_{2g} orbital system of Sr₃Ru₂O₇. -- A. P. Mackenzie's group.

• Microscopic theory with quasi-1D bands of d_{xz} and d_{yz}. Orbital degree of freedom facilitates unconventional metamagnetism exhibiting orbital ordering.

• STM quasi-particle interference as a test of orbital ordering.



• n=1: Sr₂RuO₄ p-wave superconductor with T_c=1.5K.

• n=2: Sr₃Ru₂O₇. No superconductivity. Paramagnet at B=0; **meta-magnetism** at finite B-fields.

• n>=3: ferromagnet. As $n \rightarrow \infty$ (SrRuO₃), T_c=165 K.

Metamagnetism in Sr₃Ru₂O₇

• Metamagnetism: a superlinear relation between magnetization and the B field. Analogy of FM at finite fields, but no symmetry breaking.

• Very pure samples $\rho \sim 0.4 \mu \Omega \text{ cm}$: two consecutive metamagnetic transitions at 7.8 and 8.1T from AC magnetic susceptibility (17Hz).

• Dissipative peaks in $\mbox{Im}\,\chi\,$ mean first order phase transitions (hysteresis).



Resistance anomaly

• Between two metamagnetic transitions (T<1K), the resistivity measurement shows enhanced electron scattering.

• Many measurements mark the phase boundary.

- resistivity $\partial_{H} \rho \diamond \partial_{T}^{2} \rho \blacklozenge$
- ac susceptibility
- magnetostriction
- thermal expansion △▽
- dc magnetization

Grigera et. al., Science 306, 1154 (2004)



A new phase: Pomeranchuk instability!

• Spin-dependent Fermi surface anisotropic distortion -- partly d-wave anisotropic unconventional magnetism.

• Resistivity anomaly arises from the **domain formation** due to two different patterns of the nematic states.

• Resistivity anomaly disappears as B titles from the c-axis, i.e., it is sensitive to the orientation of B-field.



Further evidence: anisotropic electron liquid

• As the B-field is tilted away from c-axis, large resistivity anisotropy is observed in the anomalous region for the inplane transport.



Borzi et. al., Science 315, 214 (2007)

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W. C. Lee and C. Wu, PRB 80, 104438 (2009). C. Wu, K. Sun, E. Fradkin, and S. C. Zhang, PRB 75, 115103 (2007).

• STM quasi-particle interference as a test of orbital ordering.

Landau Fermi liquid (FL) theory



L. Landau



• The existence of Fermi surface. Electrons close to Fermi surfaces are important → renormalized into quasi-particles.

• Interaction functions (no SO coupling):

 $f_{\alpha\beta,\gamma\delta}(\hat{p}_{1},\hat{p}_{2}) = f^{s}(\hat{p}_{1},\hat{p}_{2}) \qquad \text{density} \\ + f^{a}(\hat{p}_{1},\hat{p}_{2})\vec{\sigma}_{\alpha\beta}\cdot\vec{\sigma}_{\gamma\delta} \qquad \text{spin}$

• Landau parameter in the *I*-th partial wave channel:

$$F_l^{s,a} = N_0 f_l^{s,a} \quad N_0 : \text{DOS}$$

Pomeranchuk instability criterion



- Fermi surface: elastic membrane.
- Stability: $\Delta E_{\kappa} \propto (\delta n_{l}^{s,a})^{2}$

$$\Delta E_{\rm int} \propto \frac{F_l^{s,a}}{2l+1} (\delta n_l^{s,a})^2$$

• Surface tension vanishes at:

I. Pomeranchuk



$$F_l^{s,a} < -(2l+1)$$

- Ferromagnetism: the F_0^a channel.
- Nematic electron liquid: the F_2^s channel.

• Unconventional magnetism: F_l^a $(l \ge 1)$

The order parameters: the 2D d-wave channel

• F_2^a : spin quadrupole moments in **momentum space**.

C. Wu, K. Sun, E. Fradkin, and S. C. Zhang, Phys. Rev. B 75, 115103 (2007).

$$\vec{n}_{x^2-y^2} = \sum_{\vec{k}} \psi_k^{\dagger} \vec{\sigma} \psi_k \cos 2\theta_k$$
$$\vec{n}_{xy} = \sum_{\vec{k}} \psi_k^{\dagger} \vec{\sigma} \psi_k \sin 2\theta_k$$

- *cf.* Ferromagnetic order (s-wave): $\vec{s} = \sum_{\vec{k}} \psi_k^+ \vec{\sigma} \psi_k$
- Arbitrary partial wave channels: spin-multipole moments.

$$F_l^a: \cos 2\theta_k \to \cos l\theta_k; \sin 2\theta_k \to \sin l\theta_k$$

Mean field theory and Ginzburg-Landau free energy

• The d-wave spin exchange interaction:

$$F_2^{a} \quad H_{int} = \sum_{q} f_2^{a}(\vec{q}) \{ \vec{n}_{x^2 - y^2}(\vec{q}) \cdot \vec{n}_{x^2 - y^2}(\vec{q}) + \vec{n}_{xy}(\vec{q}) \}$$

$$H_{MF} = \sum_{k} \psi^{+}(k) [\varepsilon(k) - \mu - (\vec{n}_{x^{2} - y^{2}} \cos 2\theta_{k} + \vec{n}_{xy} \sin 2\theta_{k}) \cdot \vec{\sigma}] \psi(k)$$

• Symmetry constraints: rotation (spin, orbital), parity, time-reversal.

$$F = r(|\vec{n}_{x^2-y^2}|^2 + |\vec{n}_{xy}|^2) + v_1(|\vec{n}_{x^2-y^2}|^2 + |\vec{n}_{xy}|^2)^2 + v_2|\vec{n}_{x^2-y^2} \times \vec{n}_{xy}|^2$$

instability!

$$r = \frac{N_0}{2} \frac{1 + F_2^a / 2}{|F_2^a|} \quad F_2^a < -2$$

 β and α -phases (*d*-wave)



Ref: C. Wu, K. Sun, E. Fradkin, and S. C. Zhang, PRB 75, 115103 (2007).

New ingredients of Sr₃Ru₂O₇: t_{2g}-orbitals

• Itinerant metallic bilayer 4d-system with active t_{2g} -bands (d_{xy} , d_{xz} , d_{yz}).

• Orbitals play important roles in magnetism, superconductivity, and transport properties in transitional metal oxides.

• Orbital degeneracy and spatial anisotropy.



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Anisotropic bondings: 2D v.s. quasi 1D bands

- Longitudinal bonding: hopping assisted by oxygen; strong.
- Transverse bonding: direct overlap; weak.



• In-plane bonding. dxy-band: 2D band; dxz and dyz: quasi-1D bands.



Questions and Observations

• Q1: Generalize the unconventional magnetic states to orbital systems. Which orbital bands are responsible in Sr327?

• Q2: Landau parameters in high partial wave channels are usually not large. How to enhance the d-wave channel interactions?

• Unconventional (nematic) meta-magnetic transitions in Sr₃Ru₂O₇ are NOT observed in the monolayer compound Sr₂RuO₄.

• The bilayer splitting of the d_{xy}-band is very small. No oxygen p-orbitals are involved.

• The d_{xy}-band structures in Sr₃Ru₂O₇ and Sr₂RuO₄ are similar.



Previous theory based on the dxy-band by Kee et al.

• As the B-field increases, the Fermi surface (FS) of the majority spin expands and approaches the van Hove singularity.

• The 1st meta-magnetic transition: the FS of the majority spin is distorted to cover one of vHs along the x and y directions.

• The 2nd transition: four-fold rotational symmetry is restored.

• Drawback: an artificial d-wave channel inter-site interaction is involved.



H.-Y. Kee and Y.B. Kim, Phys. Rev. B 71, 184402 (2005); Yamase and Katanin, J. Phys. Soc. Jpn 76, 073706 (2007); C. Puetter et. al., Phys. Rev. B 76, 235112 (2007). 27

Our proposed solution: quasi-1D d_{xz} and d_{yz}

• The major difference between Sr₃Ru₂O₇ and Sr₂RuO₄ is the large bilayer splitting of d_{xz}/d_{yz} bands.

• We will see that orbital band hybridization naturally enhances the *d*-wave channel exchange interaction.



• Similar proposal has also been made by S. Raghu, S. Kivelson *et al.*, Phys. Rev. B 79, 214402(2009).

d_{xz}/d_{yz} orbital band structures and hybridizations

• For simplicity, we only keep the bilayer bonding bands of dxz and dyz. Fermi Surface in 2D Brillouin Zone



• Eigen-basis has internal *d*-wave like form factors of the orbital configurations.

Band hybridization enhanced Landau interaction in high partial-wave channels

• A heuristic argument: a hybridized band Bloch wavefunction with internal orbital configuration as

 $|\Psi_{\sigma}(p)\rangle = e^{ipr}(\cos\phi_p|d_{xz}\rangle + \sin\phi_p|d_{yz}\rangle)\otimes\chi_{\sigma}$

• The Landau interaction acquires a d-wave $d_{\rm xz}$ angular form factor as.

$$f_{\uparrow\uparrow}(\vec{p}_1, \vec{p}_2) = V(q=0) - \frac{1}{2} [1 + \cos 2\theta_{p_1 p_2}] V(p_1 - p_2)$$

$$f_{\uparrow\downarrow}(\vec{p}_1,\vec{p}_2) = V(q=0)$$

• Even V(p₁-p₂) is dominated by the *s*-wave component, the angular form factor shifts a significant part of the spectra weight into the *d*-wave channel.

VZ.

 $\theta_{p_1p_2}$

Microscopic Model

• Band Hamiltonian: σ -bonding t_{μ} , π -bonding t_{\perp} , nextnearest-neighbour hoppings t', t''

$$H_{band} = \sum_{k,\sigma} \varepsilon_x(k) d_{xz,k\sigma}^+ d_{xz,k\sigma} + \varepsilon_y(k) d_{yz,k\sigma}^+ d_{yz,k\sigma} + \lambda(k) (d_{xz,k\sigma}^+ d_{yz,k\sigma}^+ + h.c.)$$

$$\varepsilon_x(k) = -2t_{\mu} \cos k_x - 2t_{\perp} \cos k_y - 4t' \cos k_x \cos k_y$$

$$\varepsilon_y(k) = -2t_{\perp} \cos k_y - 2t_{\mu} \cos k_x - 4t' \cos k_x \cos k_y$$

$$\lambda(k) = 4t'' \cos k_x \cos k_y$$

• Hybridized eigen-basis.

$$\begin{pmatrix} \gamma_{+,k\sigma} \\ \gamma_{-,k\sigma} \end{pmatrix} = \begin{pmatrix} \cos \phi_k & \sin \phi_k \\ -\sin \phi_k & \cos \phi_k \end{pmatrix} \begin{pmatrix} d_{xz,k\sigma} \\ d_{yz,k\sigma} \end{pmatrix}, \quad \tan 2\phi_k = \frac{-4t''}{t_{//} - t_{\perp}} \frac{\cos k_x \cos k_y}{(\cos k_x - \cos k_y)}$$

$$E^{\pm}(k) = \frac{1}{2} \left(\varepsilon_{x}(k) + \varepsilon_{y}(k) \pm \sqrt{\left(\varepsilon_{x}(k) - \varepsilon_{y}(k)\right)^{2} + 4\lambda^{2}(k)} \right)$$

van Hove Singularity of density of states

 $(t_{\mu}, t_{\perp}, t', t'') = (1.0, 0.145, 0.0, 0.3)$

Mean-Field Solution based on the multiband Hubbard model

$$H_{int} + H_{zeeman} = U \sum_{i,a=xz,yz} n_{a\uparrow}(i) n_{a\downarrow}(i) + V \sum_{i} n_{xz}(i) n_{yz}(i) - J \sum_{i} \{S_{xz}(i) \cdot S_{yz}(i) - \frac{1}{4} n_{xz}(i) n_{yz}(i)\}$$
$$+ \Delta \sum_{i} d^{+}_{xz\uparrow}(i) d^{+}_{xz\downarrow}(i) d_{yz\downarrow}(i) d_{yz\uparrow}(i) + h.c. - h \sum_{a=xz,yz\sigma} \sigma d^{+}_{a\sigma} d_{a\sigma}$$

• Competing orders: magnetization(m), charge (n_c) /spin quadrupolar (n_{sp}) orders near the van Hove singularity.

$$m = \sum_{a=xz,yz} \langle S_a(z) \rangle, \quad n_c = \frac{1}{2} \{ \langle n_{xz} \rangle - \langle n_{yz} \rangle \}, \quad n_{sp} = \langle S_{xz}(z) \rangle - \langle S_{yz}(z) \rangle$$
$$H_{mf} = \sum_{k\sigma,\alpha=\pm} \xi_{\alpha\sigma} \gamma_{\alpha k\sigma}^+ \gamma_{\alpha k\sigma} + V_m m^2 + V_c n_c^2 + V_{sp} n_{sp}^2$$
$$\xi_{\alpha\sigma} = E^{\alpha}(k) - \mu - \sigma V_m m - \alpha (V_c n_c + \sigma V_{sp} n_{sp}) \cos 2\phi_k$$
$$V_m = \frac{U}{2} + \frac{J}{4} \quad , \quad V_c = V + \frac{J}{4} - \frac{U}{2} \quad , \quad V_{sp} = \frac{U}{2} - \frac{J}{4}$$

Phase diagram v.s. the B-field

• **Unconventional** metamagnetism from the **conventional** Hubbard interactions at the mean-field level.

• Nematic ordering as orbital ordering: different occupations between dxz and dyz orbitals.

Outline

• Introduction to unconventional magnetism.

• Experimental results: unconventional (nematic) metamagnetism in the t_{2g} system of Sr₃Ru₂O₇ (bilayer).

• Pomeranchuk instability of Fermi liquids.

• Microscopic theory with quasi-1D bands of d_{xz} and d_{yz}: unconventional metamagnetism with orbital ordering.

• STM quasi-particle interference as a test of orbital ordering.

W. C. Lee and C. Wu, Phys. Rev. Lett. 103, 176101 (2009);

W. C. Lee, D. Arovas, and C. Wu, Phys. Rev. B 81, 184403 (2010).

Spectroscopic Imaging STM quasi-particle interference

• Real space spectroscopy reveals Fermi surface structure. Widely used in high T_c cuprate systems.

Y. Kohsaka et al., Nature (London) 454, 1072 (2008). Q.-H. Wang and D.-H. Lee, Phys. Rev. B 67, 020511 (2003).

ARPES

-10 me

A toy model calculation: QPI of quasi-1D dxz/dyz bands

κ

• Orbital ordering can be detected by QPI.

W. C. Lee and C. Wu, PRL 103, 176101(2009).

• c.f. Ca(Fe_{1-x}Co_x)₂As₂: QPI shows nematic ordering.

Chuang et al, Science 327, 181 (2010).

2π/a_{Fe-Fe} ₽ 21,122 AS'

Band structures of Sr₃Ru₂O₇

• Complication from the orbital structure, the staggered rotation of RuO octahedra; bilayer splitting, and spin-orbit coupling.

- Fermi surfaces from the tight-binding model.
- Reduced Brillouin zone.
- Bilayer bonding (black) and anti-bonding (red) bands.

W. C. Lee, D. Arovas, and C. Wu, Phys. Rev. B 81, 184403 (2010).

Band structures of Sr₃Ru₂O₇ at surface

• Fermi surfaces measured by ARPES. Bilayer bias due to the surface effect.

A. Tamai, et al, Phys. Rev. Lett. 101, 026407 (2008)

• Tight-binding fit with V_{bias} =10% band width of d_{xz} and d_{yz} . Band crossings avoided.

STM QPI at zero field (B=0) in Sr₃Ru₂O₇

×~ 0.0

-0.5

-1.0

-1.0

0

-0.5 0.0 0.5 1.0 k_x

• Quasi-1D band structures have been seen experimentally.

J. H. Lee, et al., Nature Physics 5, 800 (2009)

• T-matrix calculation. Ring structure from quasi-1D orbital scatterings.

W. C. Lee, D. Arovas, and C. Wu, Phys. Rev. B 81, 184403 (2010).

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Summary

- Unconventional magnetism is a class of exotic states of matter.
- Quasi-1D orbital bands provide a natural explanation for the unconventional metamagnetic state observed in $Sr_3Ru_2O_7$ as orbital ordering.
- STM quasi particle interference provides a probe to orbital ordering.

