Orbital-exchange and fractional spinon excitations in an f-electron metal $\text{Yb}_2\text{Pt}_2\text{Pb}$

Igor Zaliznyak

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Contributors and collaborators

Meigan Aronson
Liusuo Wu
Bill Gannon
Alexei Tsvelik (BNL)
Jean-Sebastien Caux (U. Amsterdam)
Michael Hagemans (U. Amsterdam)
Yiming Qiu
John Copley
Jeff Lynn
Georg Ehlers, Andrey Podlesnyak, Paul Cutler
DCS/NCNR
DCS/NCNR
BT-7/NIST
CNCS/SNS/ORNL
Quasiparticles in condensed matter

Quasiparticle: phonon, magnon

\[ q = k_f - k_f \]

1 meV = 11.6 K

LaSrCuO4
The scattering experiment

$$\frac{d^2 \sigma(q, E)}{dE d\Omega} = \frac{1}{\Phi_i(k_i)} \frac{\delta I_f(k_f)}{dE_f d\Omega_f}$$

$$\eta = \eta_i E$$

$$\eta = \eta_f E$$

Figure 2-1. Typical geometry of a scattering experiment, (a) elastic, (b) inelastic.

$$\delta I_f(k_f) = \Gamma_{i \rightarrow f} \frac{d^3 k_f}{(2\pi)^3}$$

$$\Gamma_{i \rightarrow f} = \frac{2\pi}{\hbar} \left| \left\langle k_f, S_f^z, \eta_f | T | k_i, S_i^z, \eta_i \right\rangle \right|^2 \delta \left( E_i - E_f + \frac{(\hbar k_i)^2}{2m} - \frac{(\hbar k_f)^2}{2m} \right)$$

$$E = E_f(\eta_f) - E_i(\eta_i) = \frac{(\hbar k_i)^2}{2m} - \frac{(\hbar k_f)^2}{2m}$$

$$q = k_i - k_f$$
Neutron interactions and scattering cross-section: precisely known!

\[ T = V + TGV \]

Lippmann & Schwinger, Phys. Rev. 79 (1950)

\[ T = V + VGV + VGVGV + \ldots \]

\[ b = - \frac{m}{2\pi \hbar^2} T \]

\[
\frac{d^2\sigma(q, E)}{dE d\Omega} = \sum_{i,f} \frac{k_f}{k_i} \left| \langle S^z_f, \eta_f | b(-q) | S^z_i, \eta_i \rangle \right|^2 \delta \left( E_i(\eta_i) - E_f(\eta_f) + \frac{(\hbar k_i)^2}{2m} - \frac{(\hbar k_f)^2}{2m} \right)
\]

\[ b_N = b_N \delta(r_n - R) \]

\[ V_N = -\frac{m}{2\pi \hbar^2} b_N \delta(r_n - R) \]

nuclear scattering length, \( b \sim 10^{-13} \text{ cm} \)

\[
V_{mag} = -\left\{ \frac{8\pi}{3} \left( \mu_n \mu_e \right) \delta(r) - \frac{\left( \mu_n \mu_e \right)}{r^3} + \frac{3 \left( \mu_n r \right) \left( \mu_e r \right)}{r^5} \right\}
\]

magnetic scattering length, \( r_m = -5.39 \times 10^{-13} \text{ cm} \)

Strong interaction, but extremely short-range: Fermi pseudo-potential, not Born approximation!

Weak interaction: Born approximation is OK!
How to measure quasiparticle excitations: Neutron Scattering.

Long-lived quasiparticle ➔

delta function singularity in cross-section

Changes in the energy of the electrons are first used in an...

magnetic scattering length, \( r_m = \text{-}5.39 \times 10^{-13} \text{ cm} \)

Thermal, \( b \approx 10^{-13} \text{ cm} \)

Crystal that sorts and forwards neutrons of a certain wavelength (energy) – monochromated neutrons

When the neutrons penetrate the sample they start or cancel oscillations in the atoms. If the neutrons create phonons or magnons they themselves lose the energy these absorb – inelastic scattering

...and the neutrons then counted in a detector.

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How do neutrons measure excitations: triple-axis crystal spectrometer (TAS)
How do neutrons measure excitations: time-of-flight (TOF) direct geometry spectrometer (DGS)
How do neutrons measure excitations TOF DGS @SNS (ARCS)
How do neutrons measure excitations TOF DGS ARCS@SNS
How neutrons measure excitations TOF DGS SEQUOIA@SNS

Cylindrical geometry
Sample-to-detector distance
5.5–6.3 m
Incident energy range
4–2000 meV
Resolution (elastic)
1–5% $E_i$
Vertical detector coverage
-18–18°
Horizontal detector coverage
-30–60°

Simply an enormous camera streaming a movie
Example: spinons in $S=1/2$ Cu chains

Multi-spinon ($B = 0$) and magnon ($B = 5T$) excitations in copper sulphate pentahydrate
Example: spinons in S=1/2 Cu chains

Chains are formed by particular orbital overlap patterns, which define the superexchange.

Multi-spinon excitations in Sr$_2$CuO$_3$
Walters, Perring, Caux, Savici, Gu, Lee, Ku, Zaliznyak,
Nature Physics (2009)
Yb$_2$Pt$_2$Pb: f-electron Ising dimers on a Shastry-Sutherland lattice?

Kim, Benett, Aronson, PRB (2008); PRL (2013)

- Tetragonal P4$_2$/mmm: U$_2$Pt$_2$Sn type (Pöttgen 1999)
- Other Shastry – Sutherland Lattice Systems:
  - RB$_4$ (R=Pr,Gd,Tb,Dy,Ho,Er, Tm): magnetically ordered metals, Ising moments.
  - SrCu$_2$(BO$_3$)$_2$: Insulator, Cu S=1/2, spin-liquid (B=0), AF order (B>34 T).

Dimers on the Shastry-Sutherland lattice
J=intradimer coupling, J’=interdimer coupling
Dimer Liquid Magnetic Order

QCP
Excitations of a $S=1/2$ Shastry-Sutherland dimers: $\text{SrCu}_2(\text{BO}_3)_2$

- gapped triplet magnon with flat dispersion – propagates only in high order of perturbation in $J'$
- fine energy-structure from low-symmetry perturbations – DM, staggering, ...


Zayed, et. al, PRL (2012)
Yb$_2$Pt$_2$Pb crystal structure: f-electron Ising dimers on a Shastry-Sutherland lattice?

- Two Yb sites form orthogonal pairs => SSL
- 2D SSL physics: dimers, singlet ground states?
- Neutron scattering: singlet ground state - triplet magnon excitation?

M.S. Kim et al., 2008, 2012
Yb$_2$Pt$_2$Pb: f-electron Ising dimers on a Shastry-Sutherland lattice?

- Weakly coupled layers with Shastry-Sutherland lattice? Two dimensional magnetic system?
- Spin ladders with rungs in Shastry-Sutherland lattice layers? One dimensional magnetic system?

\[ d_{\text{chain}} < d_{\text{rung(red)}} < d_{\text{rung(blue)}} < d_{\text{inter-ladder}} \]

\[ J_{\text{chain}} > J_{\text{rung}} > J_{\text{inter-ladder}} \]
Yb$_2$Pt$_2$Pb CEF point charge model calculation: Ising moments

\[ \text{Yb}^{3+}: \text{CEF Levels} \]

\[ |E_{\pm}\rangle_3 = \Delta_3 \sim 94 \text{ meV} \]

\[ |E_{\pm}\rangle_2 = \Delta_2 \sim 64 \text{ meV} \]

\[ |E_{\pm}\rangle_1 = \Delta_1 \sim 37 \text{ meV} \]

\[ |E_{\pm}\rangle_0 = 0.992|\pm 7/2\rangle + 0.100|\pm 3/2\rangle + 0.064|\mp 1/2\rangle + 0.032|\mp 5/2\rangle, \]

\[ |E_{\pm}\rangle_1 = -0.082|\pm 7/2\rangle + 0.300|\pm 3/2\rangle + 0.355|\mp 1/2\rangle + 0.881|\mp 5/2\rangle, \]

\[ |E_{\pm}\rangle_2 = 0.092|\pm 7/2\rangle + 0.785|\pm 3/2\rangle + 0.420|\mp 1/2\rangle - 0.446|\mp 5/2\rangle, \]

\[ |E_{\pm}\rangle_3 = -0.153|\pm 7/2\rangle + 0.832|\pm 3/2\rangle - 0.533|\mp 1/2\rangle + 0.005|\mp 5/2\rangle, \]

\[ \text{Wu, et. al, unpublished (2015)} \]
**Yb$_2$Pt$_2$Pb: effective spin-1/2 description of the G. S. doublet**

\[ M^\alpha = g_{\text{eff}}^\alpha \mu_B S^\alpha, \quad (S^\alpha)^2 = \frac{1}{4} \]

\[ g_{\text{eff}}^z = 7.9 = g_{\text{eff}}^{(110)}(\text{Yb1}) = g_{\text{eff}}^{(-110)}(\text{Yb2}) \]

\[ g_{\text{eff}}^{x,y} \leq 0.8 \]

\[ M_{100} \approx 2.6 \mu_B, \quad M_{110} \approx 3.6 \mu_B, \quad M_{100} \approx \sqrt{2} M_{110} \gg M_{001} \]

**Yb$_2$Pt$_2$Pb: effective spin-1/2 description of the G. S. doublet**

General projected Hamiltonian:

$$H = \sum_{i \neq j, \alpha, \beta} J_{\alpha\beta} S_i^\alpha S_j^\beta - \mu_B \sum_{i, \alpha, \beta} H_{\alpha} g_{\alpha\beta} S_i^\beta, \quad S^2 = \frac{3}{4}$$

**Neutron Scattering intensity:**

$$I^{\alpha\alpha} \sim \left(1 - \frac{q_\alpha^2}{q^2}\right) \int e^{-\frac{i}{\hbar}E_t} \left\langle M_q^\alpha M_{-q}^\alpha(t) \right\rangle \frac{dt}{2\pi\hbar} \sim g_\alpha^2 \left(1 - \frac{q_\alpha^2}{q^2}\right) \int e^{-\frac{i}{\hbar}E_t} \left\langle S_q^\alpha S_{-q}^\alpha(t) \right\rangle \frac{dt}{2\pi\hbar}$$

Sensitive to the longitudinal sector only:

$$I^{zz}/I^{xx} \sim \left(\frac{g^{zz}}{g^{xx}}\right)^2 > 100$$

**Sum rule for spin-1/2:**

$$\int dE \int \frac{d^n q}{(2\pi)^n} S^{\alpha\alpha}_{\alpha\alpha}(q, E) = \frac{1}{4}$$

**Strongly Anisotropic Yb\(^{3+}\) moments: Isolated Doublet Ground State**

- **Curie Weiss Susceptibility** \(T > 300\) K: \(\chi = \chi_0 + C/(T-\theta)\)  
  \(\mu_{CW} = 4.54\) \(\mu_B/Yb\) (\(B \parallel 001, 110\))  
  \(\theta_{001} = -217\) K  
  \(\theta_{110} = 28\) K

- **Specific heat and inelastic neutron scattering** confirm CEF level spacings:  
  - Doublet ground state is energetically isolated (\(T < \sim 100\) K).

- **Point charge model:**  
  - a doublet ground state for Yb\(^{3+}\) with dominantly (\(\sim 98\%\)) \(J_z = \pm 7/2\) content  
  - an anisotropic Lande g-factor: \(g_{eff}^Z = 7.9,\) \(g_{eff}^x = 0.5\)  
  - for \(S_x = S_y = S_z\): \(M_{S(110)} = g_{eff}^Z \mu_B S_z = \pm 4\) \(\mu_B\)  
  - \(M_{S(001)} = g_{eff}^x \mu_B S_x = 0.25\) \(\mu_B\)

**Brookhaven Science Associates**  
Workshop: Beyond Integrability. Montreal, 2015
Yb$_2$Pt$_2$Pb: Ising dimers on a Shastry-Sutherland lattice?

- Strong Spin-Orbit coupling + Crystal Electric field => well-separated $J$-doublet Ground State: $|G.S.\rangle = 0.99|\pm 7/2\rangle + ...$

- Yb 4f magnetic moments, $\mu_{Yb} = g_J\mu_B J$ ($g_J=8/7$) point along (-1, 1, 0) or (1, 1, 0)

- Very strong Ising anisotropy, >30meV (> ~300 K, > ~300 T)
**Yb$_2$Pt$_2$Pb: Ising dimers on a Shastry-Sutherland lattice?**

- Broad peak in $\chi(T)$ interpreted as resulting from a gap over the non-magnetic G. S.
  - Order from from a paramagnetic liquid of Yb dimers, having a gap $\Delta \approx 4.3$ K
  - Gap $\Delta(T)$ decreases with the increasing $T$
- Mean-field-RPA expression for the $\chi(T)$ of coupled dimers:
  - $J \approx -2.3$ K, $J' \approx -1.95$ K, $J'/J \approx 0.85$, $g \approx 5.43$

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**Graphs and Data**

- Left: Plot of $\chi(T)$ with inset showing $\chi(B)$ with $B = 0.1$ T and $B \parallel [110]$
- Middle: Graphs showing $M(T)$ and $\chi(T)$ for various $T$ values.
- Right: Plot of $\Delta(T)/B$ with inset showing $T_N(K)$ and $\Delta(K)/B$.
Antiferromagnetic Order in Yb$_2$Pt$_2$Pb

- Antiferromagnetic Order in Yb$_2$Pt$_2$Pb: $T_N \approx 2.07$ K
  - Sharp cusp in specific heat $C/T$, superimposed on broad background.
  - Entropy loss $\sim 0.58 R \ln 2$.
  - Peak in $d\chi/dT$.
  - Suppression of spin disorder scattering in resistivity $\rho(T_N)$: $\rho(T=0) \sim 1.5 \mu\Omega\cdot$cm
Yb$_2$Pt$_2$Pb: Magnetization and phase diagram

- Sequence of Magnetic Plateaux phases at low temperatures
- Linear-resistivity “dome” phase in-between $B_{c1} \approx 1.25 \text{T}$ and $B_{c2} \approx 2.3 \text{T}$
- Horizontal field-independent phase lines at $T_{N1} \approx 2.07 \text{ K}$ and $T_{N2} \approx 0.8 \text{ K}$
Antiferromagnetic Order in Yb$_2$Pt$_2$Pb

- $T < T_N = 2$ K: AF supercell 5x5x1 $q_1 = (0.2, 0.2, 1)$ rlu
- Order parameter: 2D Ising universality class ($\beta = 1/8$)
  
  $I \propto (T - T_N)^{2\beta} \quad \beta = 0.12$
  $I \propto (B - B_{QCP})^{2\beta}, \beta = 0.15$

7 g array of ~400 aligned crystals.
Antiferromagnetic Order in Yb$_2$Pt$_2$Pb

- Yb$_2$Pt$_2$Pb: B=0, two orthogonal AF propagation wave vectors: $\mathbf{q}_{1,2}=(\pm 0.2,0.2,0)$ ru in $ab$- (SSL) plane
- $B \parallel (-110)$: only the $\mathbf{q}_2=(-0.2,0.2,0)$ sublattice survives, $\mathbf{q}_1=(0.2,0.2,0)$ sublattice is fully polarized.
- Ising character: Yb moments can be either parallel or perpendicular to (110),(-110) dimer bonds but magnetic structure has moments perpendicular to dimers.
  - Sublattice $\mathbf{q}_1=(0.2,0.2,0)$: dimer bonds $\parallel$ (110), moments along (-110)
  - Sublattice $\mathbf{q}_2=(-0.2,0.2,0)$: dimer bonds $\parallel$ (-110), moments along (110)
Antiferromagnetic Order in Yb$_2$Pt$_2$Pb

\[- \frac{\mu_0}{4\pi} \frac{1}{r^3} (3(m_1 \cdot r)(m_2 \cdot r) - (m_1 \cdot m_2))\]

- $\mathbf{q}_1 = (0.2,0.2)$ 5x5 ordering in the a-b plane minimizes the energy of dipole-dipole interactions!
  - Yb(1) and Yb(2) dimer sublattices are decoupled
Magnetic order in Yb$_2$Pt$_2$Pb in magnetic field: magnetization and incommensurability

\[ \Delta q = m: \text{magnetization in spin-1/2 1D chain?} \]
Yb$_2$Pt$_2$Pb: magnetic excitations

- Dispersing excitations along (00L)
  - continuum of fractional quantum spinons
- Gapped, dispersion-less excitations in the HHL (SSL) plane
  - excitations are one-dimensional, propagation confined to c-axis
Orbitals and emerging one-dimensionality in $Yb_2Pt_2Pb$

- $J$ is dominated by $L = 3$ orbital angular momentum, spin is “quenched” by spin-orbit
- Orbital overlaps generate one-dimensional pattern
- Six-fold symmetry of the f-orbital makes ladder legs and rungs inequivalent
- Chains, not ladders
- Orbital spinons, charge-orbital separation, …
Magnetic excitations in Yb$_2$Pt$_2$Pb: suppression by magnetic field

At $B = 4$T one sublattice is fully polarized; the other is not affected

- the energy integrated spectral weight is half that for $B = 0$
- no coherent magnon/spin wave!
- seeing the longitudinal sector only: magnon is dark matter!
Polarization of magnetic excitations in Yb$_2$Pt$_2$Pb: longitudinal dynamics

- Sensitive to longitudinal fluctuations only
  - no transverse components are observable
  - no spin waves seen
Polarization of magnetic excitations in Yb$_2$Pt$_2$Pb: longitudinal dynamics

- B=4T, sublattice Yb(2) with moments along (1, 1, 0)
  - Polarization factor for Yb(2) sublattice:

- B=0-4T, sublattice Yb(1) with moments along (-1, 1, 0)
  - Polarization factor for Yb(1) sublattice is constant: $P = 1$

$$P = \frac{Q_l^2}{Q_{hh}^2 + Q_l^2}$$
Magnetic excitations in Yb$_2$Pt$_2$Pb:
onset of the continuum

A

\[ Q_{00L} = [0.45, 0.55] \]
\[ Q_{HH} = [0, 1] \]

B

\[ Q_{00L} = [0.95, 1.05] \]
\[ Q_{HH} = [0, 1] \]

C

\[ E = [0.15, 1.5] \text{ meV} \]
\[ \Delta = 2.6 \]
\[ \Delta = 3.46 \]

D

\[ M(Q) / \mu_B \text{ / Yb} \]
\[ O0L \text{ (r.l.u.)} \]
Magnetic excitations in Yb$_2$Pt$_2$Pb: comparison with theory (JSC)

Effective spin-1/2 Hamiltonian:

$$H_{XXZ} = J_c \sum_n \left( S_n^x S_{n+1}^x + S_n^y S_{n+1}^y + \Delta S_n^z S_{n+1}^z \right)$$

- Best match of the spectral weight:
  - $J \approx 0.2$ meV, $\Delta \approx 2.6$

- Best fit of the lower boundary:
  - $J \approx 0.1$ meV, $\Delta \approx 3.46$

- Large spectral weight at high energy, above 2-spinon continuum
  - 4-spinon continuum enhanced
Orbital-exchange and four-spinon excitations

**D**
- $S^z = \pm 1/2$
- $\Delta S^z = \pm 1$

- Interaction with neutron causes one spin to flip, creating two spinons
- Two-spin flips move two spinons apart

**E**
- $J^z = \pm 7/2$
- $\Delta J^z = 0$

- Two spinons are created when electrons hop, exchanging their orbitals
- Further-neighbor electron hopping creates 4 spinons
Magnetic excitations in Yb$_2$Pt$_2$Pb: temperature dependence

- Susceptibility is accounted for
- Sum rule holds
Emergent dynamics of $J_z = \pm 7/2$ doublets manifold

$$H = -\sum_{i,j} \sum_{m=-L}^{L} \sum_{\sigma=\pm 1/2} t(m) f_{im\sigma}^* f_{jm\sigma} + H_{corr}$$

$$H_{XXZ} = J_c \sum_n \left( \Delta S^z_n S^z_{n+1} + S^x_n S^x_{n+1} + S^y_n S^y_{n+1} \right)$$

$J_{eff} = t^2/U$

- Virtual CEF excitations (e.g., $Li_{1-x}Ho_xF_4$, $RTi_2O_7$): ground state doublet is nearly pure $J_z = \pm 7/2$
- Correlated hopping via conduction electrons: nearly isotropic coupling described by XXZ Hamiltonian (Uimin 2000)
Summary

- Quantum spin-1/2 dynamics emerges in a system of mainly orbital magnetic moments in Yb$_2$Pt$_2$Pb
  - dynamics rendered by electron hopping – orbital exchange mechanism: $S^+ S^- \sim (J^+)^7 (J^-)^7$
  - orbital spinons, charge-orbital separation
  - large J is not classical

- Transverse fluctuations of the effective spin-1/2 are unobservable – “hidden”
  - unique probe of the longitudinal dynamics of the effective model
  - mechanism for hidden orders – URu$_2$Si$_2$, …
  - dark matter (hidden magnons), dark energy

- 4-spinon states result from long-range interactions (hopping)
  - Theory?