Hybrid nuclear-electronic order at 2 mK in the heavy fermion system YbRh₂Si₂

- Recent observation in YbRh₂Si₂:
- → at 2 mk: magnetic transition combined with onset of superconductivity
- Very unusual, mechanism unclear,
 - proposed to be hybrid nuclear-electronic order
- My own ideas on the origin of the 2 mK transition in YbRh₂Si₂

How can tiny nuclear moments induce a new electronic state in a lattice of strongly interacting, large moment Kramers ions

- Discussion with and strong support from Manuel Brando + Alexander Steppke (MPI CPfS)
- Interesting and constructive discussion with Q. Si, E. Schubert, and F. Steglich

The heavy fermion system YbRh₂Si₂

YbRh₂Si₂: Prominent system in the field of QCP

- Heavy Fermion system, Kondo scale $T_K \approx 20$ K
 - * AFM order at very low T: $T_N = 70 \text{ mK}$,
 - \rightarrow very close to the AFM QCP
 - * QCP induced by small magnetic field or small negative pressure



M. Brando et al., Phys. Stat. Sol. 3 (2013) 485



AFM order at $T_N = 70 \text{ mK}$

- Large λ -type anomaly in C(T) at 70 mK
 - entropy: $S(T_N) \cong 3\%$ Rln2 (Kondo screening)
- In-plane field (strong XY anisotropy):
- Peak in $\chi(T)$ at T_{N}
- Kink in magnetization at B_c = 0.06 T
- \bullet Size of ordered moment: – $\mu SR:$ 0.002 μ_B
- AFM structure still unknown

New transition at 2 mK

- E. Schubert et al., Science 351 (2016) 485
- Measurements:
 - dc magnetization with SQUID
 - ac susceptibility with SQUID and with conventional coil technique
- \rightarrow clear and sharp anomaly at T_{HYB} = 2 mK
- \rightarrow opening of a hysteresis below T_B = 10 mK
- → zfc signal at very low field diamagnetic
- \rightarrow strong suppression with increasing field





- \Rightarrow clear evidence for a magnetic transition at T_{HYB}
- ⇒ preliminary evidence for onset of superconductivity
- Superconductivity confirmed in recent noise resistivity measurements by J. Saunders et al.

Initially proposed scenario

Scenario proposed in E. Schuberth et al.:

- At 2 mK onset of hybrid nuclear/electronic order
 - \rightarrow suppress 4f AFM magnetic order
 - \rightarrow results in crossing QCP towards the PM regime
 - → induces superconductivity
- Nuclear order parameter: primary order parameter of hybrid order
 - electronic component of hybrid order undefined
- Analyzed using 3 components Landau type model
- → 9 parameters: 1 parameter fixed: hyperfine coupling - 2 constrains: T_{AF} and T_{HYB}



⇒ Huge number of free parameters: - parameters can be tuned to support proposed scenario - but many other scenario can be modelled as well

- \Rightarrow Ad hoc scenario! very speculative model has only limited significance
- only weak connection to properties of YbRh₂Si₂ some problematic assumptions
- \Rightarrow met strong skepticism

Need for a more stringent approach with a better connection to specific properties of YbRh₂Si₂

Here: alternative approach: let's start from YbRh₂Si₂

- Empirical approach
- discuss hyperfine coupling in Yb
- Single out very peculiar properties of YbRh₂Si₂
- \rightarrow Because of these peculiar properties, nuclear moments are expected to interfere with electronic moments at T ≈ 2 mK
- \Rightarrow New model: coupled nuclear electronic transition
 - but 4f moments increase below T_{HYB} instead of decreasing
- Analyze possible origin for peculiar properties of YbRh₂Si₂

Nuclear versus electronic entropy

Electronic entropy

- Analysis of C(T, B) data
 - → Reliable map of Entropy S(T,B)
- \rightarrow reasonable approximation below 30 mK

 \Rightarrow At 30 mK S_{elec} = 0.01 Rln2

 \Rightarrow At 3 mK S_{elec} = 0.001 Rln2

Yb nuclear entropy

- 171 Yb I = 1/2 abundance 14 % 173 Yb I = 5/2 abundance 16 %
- Analysis of C(T) data
 + Estimation of quadrupolar splitting
 → Yb-Nuclear Entropy: S_{nuc-Yb}(T)
 - \Rightarrow At 30 mK S_{nuc-Yb} = 0.56 Rln2

 \Rightarrow At 3 mK S_{nuc-Yb} \cong 0.5 Rln2

- \Rightarrow At 3 mK: Yb-nuclear entropy >>> electronic entropy
- Strong coupling between Yb-nuclear moment and electronic Yb 4f moments
- → any change in 4f states shall affect splitting of Yb-nuclear magnetic levels
- 2 mK transition: anomaly in $\chi(T) \rightarrow$ 4f moments are affected \rightarrow nuclear moments affected too
- ⇒ Regarding entropy: 2 mK transition is completely dominated by nuclear contribution
- \Rightarrow Nuclear moments are strongly involved in 2 mK transition

Hyperfine coupling in Yb

- Magnetic rare earth elements: very strong hyperfine coupling
- Hyperfine coupling in Yb completely dominated by effect of 4f-shell
- Two different regimes: "fast" and "slow" relaxation regimes



• YbRh₂Si₂: strong Kondo interaction \rightarrow "fast relaxation regime" - confirmed by experiments \Rightarrow 4f moments arrange according to RKKY and Kondo interactions (don't care about nuclear m) \Rightarrow Yb nuclear moment arrange according to static dipolar + quadrupolar field produced by 4f el.

Hyperfine coupling in fast relaxation regime

 \Rightarrow Nuclear moment in dipolar hyperfine field H_{hyp}

- $H_{hyp} \propto$ on-site static 4f polarization m_{4f} : H_{hyp} = A * m_{4f}
- with A \cong 110 T/ μ_B (theory + experiment)
- Example YbRh₂Si₂ (T < 200 mK):

Apply a small external in-plane field



ightarrow H_{hyp} factor 170 larger than H_{ext} \Rightarrow only on-site static 4f polarization matters

Problems for nuclear order induced transition

Fast relaxation regime: → 4f moments arrange according to electronic interactions
 → 4f moments define state of nuclear moments, not the inverse

- Further problem for nuclear order on its own
 - only 32% of Yb nuclei carry a nuclear moment
 - \rightarrow 68 % of Yb nuclei inactive
 - Two different types of nuclear moments: different I, different moment size

- coupling of I and J: AFM in ¹⁷¹Yb, FM in ¹⁷³Yb

- ⇒ 32 % active nuclear moments, with two different kinds of properties, are statistically distributed in the lattice
- \Rightarrow Nuclear order on its own would certainly be a short range, spin glass type \rightarrow No sharp transition
- But regarding entropy nuclear moments are dominant
- How to overcome this problem?

 \rightarrow Electronic order is primary order parameter of hybrid state

- In-plane magnetization of YbRh₂Si₂
 - M(H) increases linearly with H until $H = H_c$:
 - \rightarrow as expected for a standard AFM
 - ordered moments rotate towards external field

Low field in-plane magnetization of YbRh₂Si₂ *M. Brando et al., Phys. Stat. Sol. 3 (2013) 485*



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- ! YbRh₂Si₂: nuclear specific heat: local static moment decreases strongly below H_c!

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Comparison: bulk magnetization and m_{4f} deduced from $C_{nuclear}$ YbRh₂(Si_{0.95}Ge_{0.05})₂ 0 0 0 0 0 0 0.5 B (T)





- H < H_c: m_{4f} follows bulk magnetization M(H)
- µSR results: K. Ishida et al., 2003
- \Rightarrow T < T_N: increase in relaxation rate Λ very small
- \Rightarrow tiny ordered moment $m_{AFM}(H$ = 0) \approx 0.002 μ_B much smaller than saturation moment

Looking at energies: a first clue

Extremely small AFM ordered moment: $m_{AFM}(B=0) \approx 0.002 \ \mu_B/Yb$

Tiny field $B_c = 0.07$ T sufficient to induce much larger saturation moment $m_{sat} = 0.10 \mu_B/Yb$

→ Energy required to induce a larger moment is very small: $\Delta Esat_{mag} = 1/2 * m_{sat} * B_c$ $\Rightarrow \Delta Esat_{mag}/k_B = 2.3 \text{ mK}$

Increase in static local moment → increase in nuclear polarization energy

$$\begin{split} \Delta \text{Esat}_{\text{nuclear}} = n * \Delta \text{H}_{\text{hyp}} * \text{m}_{\text{nucl}} & n = 0.30 : \text{proportion of Yb with a nuclear moment} \\ \Delta \text{H}_{\text{hyp}} = \text{A}_{\text{hyp}} * \Delta \text{m}_{\text{sat}} = 11 \text{ T} \\ \text{m}_{\text{nuclear}} = 0.6 \ \mu_{\text{N}}/\text{Yb} : \text{ Mean Value (}^{171}\text{Yb} + ^{173}\text{Yb}\text{)} \\ \Rightarrow \Delta \text{Esat}_{\text{nuclear}}/\text{k}_{\text{B}} = 0.72 \text{ mK} \end{split}$$

 \Rightarrow Gain in nuclear polarization energy: same order of magnitude as 4f polarization energy

Can nuclear polarization energy replace external field?

- Energy required to increase the electronic static local polarization: $\propto (m_{4f})^2$
- Gain in nuclear polarization energy: $H_{HYP} \propto m_{4f} \longrightarrow \Delta E_{nuclear} = \propto m_{4f}$
- \Rightarrow At low T nuclear polarization energy wins

- How large shall be m_{4f} induced by hyperfine coupling at T = 0 ?
- simple approximation using measured magnetization and hyperfine coupling
- ⇒ induced moment: $m \cong 0.02 \ \mu_{\rm B}/{\rm Yb}$: a factor of 10 larger than ordered moment at 30 mK!
- ⇒ At T = 0, ignoring further possibilities, hyperfine coupling would lead to an increase of the ordered moment by one order of magnitude!
- ⇒ in YbRh₂Si₂, AFM state with very low ordered moment is inherently unstable at very low T against an increase of the 4f moment due to nuclear polarization!



Instability towards larger moment visible in low T susceptibility

• Simple thermodynamic model: 4f moments + hyperfine coupled nuclear moments

- Free Energy: $G(T, B) = G_{el}(T, B) + G_{nucl}(T, H_{hyp}) \rightarrow M(T) = \partial G / \partial B$
- $\rightarrow \chi(T) = \chi_{4f-0} + 1 / T * K * (\chi_{4f-0})^2$ $\chi_{4f-0} : 4 \text{ f susceptibility without nuclear coupling}$ $K = n * (A_{hyp} * m_{nuc})^2$

 \Rightarrow At low T, 4f polarization enhanced due to hyperfine coupling \rightarrow Curie-Weiss upturn in χ (T)

same result as for hyperfine enhanced Susceptibility of YbRh₂Si₂ at low T Van Vleck paramagnets 10 YbRh₂Si₂ H⊥c No free parameter ! *M/B* (10⁻⁶ m³/mol) - χ_{4f-0} fixed by $\chi(T)$ above 10 mK $\mu_0 H = 10 \text{ mT}$ \rightarrow Reproduces very nicely increase in $\chi(T)$ for $T_{HYB} < T < 30$ mK 8 $\chi_{hvb} = 8.0 \cdot 10^{-6} \,\mathrm{m^3/mol} \cdot (1 + 0.30 \,\mathrm{mK}/T)$ \Rightarrow Direct evidence for instability towards states with larger 10 100 ordered 4f moment *T* (mK

Energy landscape of electronic magnetic states in YbRh₂Si₂

- Magnetization + specific heat \rightarrow free energy of some relevant states
- Energy in k_{b} , zero is "T = 0" paramagnetic state
- Small moment AFM state (SM-AFM) formed at 70 mK \rightarrow Condensation energy $\Delta G \cong$ 0.1 mK
- FM state with "large" ordered moment = 0.1 μ B $\rightarrow \Delta G \cong$ + 2.2 mK
- In YbRh₂Si₂: AFM order more stable than FM order \rightarrow likely an AFM state with large ordered moment $m_{4f} \approx 0.1 \mu_B$ (LM-AFM) in-between FM and SM-AFM state



- Transition temperature?
- \Rightarrow strongly defined by achievable nuclear Zeeman splitting Δ_{ZM}
- \rightarrow T has to be sufficiently low to only populate lowest nuclear Zeeman level

 $\rightarrow m_{4f} \approx 0.1 \ \mu_B \rightarrow \Delta_{ZM} = 4.8 \ mK$ \Rightarrow T_{hyb} of the order of 1 mK



Possible origin for tiny AFM ordered moment and for peculiar field dependence of m_{4f}

- Crystal electric (CEF) in YbRh₂Si₂ \rightarrow huge anisotropy, almost pure XY behavior
- CEF ground state doublet: very large in-plane moment m_{ab} = 1.8 μ_B
 - tiny z-axis moment $m_c \leq 0.08 \ \mu_B$
- ⇒ General belief: AFM ordered moment oriented in-plane (but no experimental proof!)



But very surprising results in the alloy YbRh₂Si₂ - YbCo₂Si₂

• YbCo₂Si₂ : ($T_K \ll T_{RKKY}$) \rightarrow stable local 4f moment system

- AFM state below $T_N = 1.7$ K: large ordered moment oriented in-plane (as expected)
- But for 0.25 < x < 0.6:
 FM state ordered moment along c axis (a mystery!)
- B-T phase diagram for x < 0.25: \rightarrow c component also close to pure YbRh₂Si₂

AFM moment along c in pure YbRh₂Si₂ too?

- already suggested in PhD Thesis of Jeroen Custers (TU Dresden, 2004)
- → would explain tiny size of ordered moment:
- Size of Kondo screening? m_{sat} = 0.12 μ_B in a-b-plane factor 15 smaller than in-plane CEF m_{sat}
 - → Kondo screening reduces CEF moment by a factor 15
 - compatible with entropy $S(T_N) = 0.03 \text{ Rln2}$

⇒ moment along c direction expected to be reduced by a factor 15 too

 $\rightarrow m_c \,{\leq}\, 0.08 \; \mu_B \;$ /15 $\; {\leq}\, 0.005 \; \mu_B$

\Rightarrow Provides simple explanation for peculiar field dependence of m_{4f}

- Increase of local static moment $m_{4f}(B)$ for in-plane field:
 - → rotation from c-axis (small moment) to a-b-plane (large moment)
- Then very likely that
 - AFM state with large in-plane ordered moment is close in energy to - AFM ground state with small c ordered moment



suggests following scenario for 2mK transition

- Below T_N = 70 mK, AFM moments order along c
- Transition at 2 mK: upon cooling, transition from a - c oriented, small moment AFM state
 - to a in-plane oriented large moment AFM state
 - stabilized by the larger nuclear polarization
- → Superconductivity not connected with AFM-PM QCP
 - but with critical point associated to a change of magnetic structure
- relation to reentrant superconductivity in URhGe?
 - Large field along hard axis
 - → Moments switch toward hard axis
 - \rightarrow Superconductivity with enhanced Tc





F. Lévy et al., Science 309 (2005) 1343

Field induced Kondo breakdown in YbRh₂Si₂ ???

- Theoretical calculations for Kondo breakdown scenario:
 - always as a function of hybridization
 - no calculation as a function of magnetic field H
- In presentation/papers:
 - suggestion: $H \cong$ increasing hybridization
 - ⇒ A priory wrong! magnetic field drives Kondo systems towards localized state
 - ⇒ In general: H ≅ decreasing hybridization
- → Claim for field induced Kondo breakdown needs some additional mechanism



- e.g. like FM Ising system in a transverse field: → increasing H reduces static polarization
 - yet no proposition for an appropriate mechanism in YbRh₂Si₂

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- e.g. like FM Ising system in a transverse field: → increasing H reduces static polarization
 - yet no proposition for an appropriate mechanism in YbRh₂Si₂
- YbRh₂Si₂: nuclear specific heat ⇒ static 4f polarization continuously increases with in-plane H !
- same as for Co doping or isostatic pressure: → in-plane H ≅ decreasing hybridization
- \Rightarrow in-plane magnetic field continuously shift YbRh₂Si₂ towards a polarized localized state
- ⇒ very strongly question the possibility for a field-induced Kondo breakdown
- \Rightarrow no sound theoretical basis for claiming a field induced Kondo breakdown in YbRh₂Si₂ (H II ab)

Summary

- YbRh₂Si₂ \rightarrow two very peculiar properties of AFM state formed at T_N = 70 mK:
 - an extremely small value of the ordered moment $\,\,m_{4f}\,_{AFM}\approx 0.002\,\,\mu_B$
 - tiny external field is sufficient to induce a much larger local moment $m_{4f} \approx 0.10 \ \mu_B$
- \Rightarrow both properties makes this AFM state inherently unstable against a large moment state stabilized by nuclear polarization at $\approx 1 \text{ mK}$
- ⇒ 2 mK transition: transition from a small ordered moment AFM state to - a large ordered moment AFM state
 - stabilized by nuclear polarization energy

2 mK transition: transition to a smaller value of the ordered 4f moment?

- Hyperfine coupling: YbRh₂Si₂ is in the fast relaxation regime
- \rightarrow problem reduces to nuclear moment in hyperfine field produced by 4 f moment
- \rightarrow equivalent to paramagnetic moment in external field
- Entropy S(B, T) of PM moment in external field: universal function of T/B, \rightarrow monotonously increasing with T/B
- For nuclear system: B = $H_{HYP} \propto m_{4f}$

 \rightarrow Entropy of nuclear system is a function of T/m_{4f}, monotonously increasing with T/m_{4f}

- Because $S_{nuclear}(T) >>> S_{elec}(T) \rightarrow S_{nuclear}$ has to decrease below $T_{HYB} = 2 \text{ mK}$
- \rightarrow therefore T/m_{4f} has to decrease below T_{HYB}
- → m_{4f} cannot decrease faster than T
- Experimental evidence for a peak in C(T) at T_{HYB} , \rightarrow significant reduction of S_{total} $\Rightarrow m_{4f}$ has to increase below T_{HYP}
- \Rightarrow Transition at 2 mK cannot be associated with a transition from a large (T > T_{HYB}) to a small (T < T_{HYB}) 4f ordered moment AFM state

Slow or fast relaxation regime in Mößbauer spectroscopy

- Mößbauer Spectroscopy: active isotope is ¹⁷⁰Yb
- Ground state: I = 0,
- but excited state at 84 keV: I = 2
- Case A): "fast relaxation"
 - \rightarrow I = 2 system in an external field
 - → in paramagnetic case only quadrupol field
 - quadrupol field depends on CEF ground state
- Case B): "slow relaxation"
 → F = I + J = 2 + 1/2 = 5/2 (J = 1/2: CEF ground state doublet)
- → Hyperfine coupling splits F multiplett (even at B = 0)
 - splitting dependent on CEF ground state

 \Rightarrow Slow or fast: very different spectra

Calculated Mößbauer spectra for paramagnetic Yb and large CEF anisotropy

I. Nowik, S. Ofer, J. Phys. Chem. Solids 29 (1968) 2117



Mößbauer on YbRh₂Si₂ : fast relaxation regime

- Slow or fast: very different Mößbauer spectra
- Experiments: G. Knebel et al.,
- \Rightarrow Single line, slightly broadened
- J. Plessel et al., PRB 67 (2003) 18403
- \Rightarrow Similar result at 1.3 K

Calculation for slow relaxation regime with CEF of YbRh₂Si₂ (G. Knebel et al.)





- \Rightarrow Exp. results incompatible with slow relaxation regime
- \Rightarrow Exp. results in agreement with fast relaxation regime
 - Quadrupol moment of Γ^7 CEF ground state proposed for ${\rm YbRh_2Si_2}$ very small
 - → Expected quadrupole splitting < natural line width
 → single line expected

Fast relaxation regime expected from large Kondo scale $T_K \cong 20 \ K$

Small or large Fermi surface in YbRh₂Si₂?

Our ARPES results (K. Kummer et al., PRX 5, 011028 (2015)
 → Large fermi surface at B = 0 for 1 K < T < 100 K

→ strong controversy: - some parts of the community: cannot be !
 - other parts of the community: as expected !

- Recent STM results (S. Seiro, S. Wirth, unpublished, presented by S. Wirth at the ICM 2015)
 ⇒ at 300 mK, no significant changes in STM spectra for 0 < H < 9 T (across H*)
- general consensus: YbRh₂Si₂: at H > H* \rightarrow large FS
- same STM spectra at H = 0 and at H = 9 T >> H*
 → large FS at T = 300 mK, B= 0 too
 - → confirms our conclusions from ARPES
- ⇒ Large FS is present in a large H-T region completely enclosing the AFM state
- ⇒ In disagreement with original phase diagram drawn for Kondo breakdown scenario
- ⇒ supports SDW scenario



Energy landscape E(m_{ordered}, Q_{ordered}) in YbRh₂Si₂ b) paramagnetic versus AFM state

- Energy difference between AFM and PM state? = condensation energy of AFM state
- Can be estimated from specific heat data: $\rightarrow \Delta G = -\iint_0^{TN} (C_{AFM}(T) C_{PM}(T))/T * dT^2$
- C_{AFM}(T): experimental data
- C_{PM}(T): reasonable estimation using conservation of entropy at T_N and results for 5 % Ge-doped YbRh₂Si₂
- $\rightarrow \varDelta G = f * T_N^2 * (\varDelta C_{MF}/T_N)$
 - ΔC_{MF} : mean field step in C(T) at T_N
 - f depends on T dependence of C_{AFM}
 - YbRh₂Si₂: $C_{AFM}(T)/T \approx T^2$ $\rightarrow f \cong 0.1$
- $\Rightarrow \Delta G \approx 0.9 \text{ mJ/(K.mol)}$
- $\Rightarrow \Delta G/R \approx 0.1 \text{ mK}$

Specific heat of pure and 5 % Ge doped YbRh₂Si₂



 \Rightarrow Kondensation energy of AFM state is of the order of 0.1 mK $* k_B$

Magnetic order in YbRh₂Si₂ - YbCo₂Si₂ system



Energy landscape of magnetic states in YbRh₂Si₂

- Small $\Delta E_{mag} \rightarrow$ (large moment) FM state close in Energy to (low moment) AFM state
- Consider hypothetical FM state with $m_{FM} = m_{sat}(B_c) = 0.10 \ \mu_B/Yb$
- Calculation of differences in electr. free energy: standard approach used for superconductors
- \Rightarrow At B = 0, T = 0: $(G_{FM} G_{AFM})/k_B = 2.3 \text{ mK}$
- \Rightarrow FM state with "large" m_{4f} just 2.3 mK above AFM state with small m_{4f}
- Energy difference between AFM and PM state? = condensation energy of AFM state
- Can be estimated from specific heat data: $\rightarrow \Delta G = -\iint_0^{TN} (C_{AFM}(T) C_{PM}(T))/T * dT^2$ \Rightarrow At B = 0, T = 0: $(G_{PM} - G_{AFM})/k_B \approx 0.12 \text{ mK}$
- \Rightarrow Kondensation energy of AFM state is of the order of 0.1 mK

Energy landscape of magnetic states in YbRh₂Si₂: very small energy scale comparable to accessible gain in nuclear polarization energy