Anomalous metamagnetism in the low carrier density Kondo lattice YbRh$_3$Si$_7$

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I. CRYSTALLOGRAPHIC ANALYSIS

Figure S1 shows the crystallographic unit cell and powder x-ray diffraction at room temperature. Table S1 shows the crystallographic data for as-grown and 150 hours annealed samples, with the atomic parameters of the latter given in Table S2.

Table S1: Crystallographic data for 150 hours annealed and as grown single crystals of YbRh$_3$Si$_7$ at $T = 298$ K (space group R3c)

| formula | as grown | annealed (150 hrs) |
|---------|----------|-------------------|
| $a$ (Å) | 7.5458(4) | 7.5482(4)        |
| $c$ (Å) | 19.8240(11) | 19.8234(11)     |
| $V$ (Å$^3$) | 977.54(12) | 978.13(12)     |
| absorption coefficient (mm$^{-1}$) | 22.86 | 15.23          |
| measured reflections | 7368 | 7697          |
| independent reflections | 321 | 319            |
| $R_{int}$ | 0.063 | 0.039         |
| goodness-of-fit on $F^2$ | 1.33 | 1.26          |
| $R_1(F)$ for $F_o^2>2\sigma(F_o^2)$ | 0.018 | 0.011        |
| $wR_2(F_o^2)$ | 0.046 | 0.023        |
| extinction coefficient | 0.054(4) | 0.00245(17) |

$^a R_1 = \sum || F_o |-| F_c ||/ \sum | F_o |$

$^b wR_2 = \frac{\sum [w(F_o^2 - F_c^2)]^2}{\sum [w(F_o^2)]^2}^{1/2}$

II. COMPARISON OF TEMPERATURE DEPENDENCE OF RESISTIVITY BETWEEN A SEMIMETAL AND A DENSE KONDO LATTICE

A continuous increase of $\rho$ with decreasing $T$ through a maximum of $\rho$ at low $T$ is not only a signature for a dense Kondo lattice, but has also been observed in magnetic semimetals without Kondo effect. Figure S2(a) shows quadratic temperature dependence of $\rho$ at low temperatures of YbRh$_3$Si$_7$ at different annealing times. Below 2 K, the data are well described by $\rho = \rho_0 + AT^2$, which is typical for dense Kondo lattice compounds. Imperfection in crystals, such as grain boundaries, disorder, or the presence of impurities, gives rise to a residual resistivity $\rho_0$. In our as-grown YbRh$_3$Si$_7$ single crystals, since x-ray diffraction data does not reveal any traceable amount of impurities, $\rho_0$ is mainly due to grain boundaries, and disorder. We plot $\rho_0$ vs. annealing time in the inset of Fig. S2(a). Continuous
TABLE S2: Atomic positions, site symmetry, $U_{eq}$ values, and occupancies for 150 hours annealed and as grown single crystals of YbRh$_3$Si$_7$.

| Atom   | Site symmetry | x       | y       | z       | $U_{eq}$ (Å$^2$) | Occupancy |
|--------|---------------|---------|---------|---------|-----------------|-----------|
| as grown |               |         |         |         |                 |           |
| Yb     | $\bar{3}$     | 0       | 0       | 0       | 0.00387(19)     | 1         |
| Rh     | $\bar{2}$     | 0.32060(5) | 0       | $\frac{1}{4}$ | 0.0018(2)     | 1         |
| Si1    | 1             | 0.53733(13) | 0.67865(14) | 0.02972(4) | 0.0042(4)     | 1         |
| Si2    | 3 2           | 0       | 0       | $\frac{1}{4}$ | 0.0041(10)    | 1         |
| annealed |               |         |         |         |                 |           |
| Yb     | $\bar{3}$     | 0       | 0       | 0       | 0.00402(9)      | 1         |
| Rh     | $\bar{2}$     | 0.32063(3) | 0       | $\frac{1}{4}$ | 0.00225(11)   | 1         |
| Si1    | 1             | 0.53739(10) | 0.67873(10) | 0.02973(3) | 0.0044(2)     | 1         |
| Si2    | 3 2           | 0       | 0       | $\frac{1}{4}$ | 0.0034(5)     | 1         |

$U_{eq}$ is defined as one-third of the trace of the orthogonalized $U_{ij}$ tensor.

FIG. S1: The crystal structure of YbRh$_3$Si$_7$ in the hexagonal lattice setting. (a) Face-sharing, nearly one-dimensional Rh octahedra (green) with corner-sharing Si double tetrahedra (pink) and Yb atoms (blue). (b) The Yb sublattice in one unit cell. (c) Room temperature powder x-ray diffraction pattern for YbRh$_3$Si$_7$ (black symbols) together with the calculated pattern (red line) for space group $R\bar{3}c$ and lattice parameters $a = 7.5482(4)$ Å and $c = 19.8234(11)$ Å. Inset: two-dimensional view of the crystal structure, viewed down the c axis. All listed lattice parameters in the text and crystallographic directions correspond to the hexagonal-equivalent unit cell.
FIG. S2: (a) Quadratic temperature dependence of $\rho$ of YbRh$_3$Si$_7$ at low temperatures and for different annealing times. Black solid lines represent fits of $\rho = \rho_0 + AT^2$. The inset shows $\rho_0$ vs. annealing time. (b) Laue x-ray pattern taken along the hexagonal [00-1] direction.

suppression of $\rho_0$ by increasing the annealing time validates that imperfection in crystals is largely minimized in the optimally-annealed sample.

We show a Laue picture of optimally-annealed YbRh$_3$Si$_7$ crystals in Fig. S2(b), where the black points are measured, and red are calculated. The measured reflections are remarkably sharp, even at large angles. These are reproducible measurements, with minimal differences between > 5 different measured points/sample for two different annealed samples. The sharp reflections, together with the agreement between measurements and calculations, as well as single crystal data probing the composition as exactly 1:3:7 are strong arguments for high crystal quality.

A continuous increase of $\rho$ with decreasing $T$ through a maximum of $\rho$ at $T^*$ is not only a signature for a dense Kondo lattice, but has also been observed in magnetic semimetals/semiconductors without Kondo effect. For the latter, the rising part of resistivity can be described by $\rho \sim \exp(\Delta/k_B T)$, where $\Delta$ is the activation energy. Figure S3(a) shows that the resistivity of the as-grown and optimally annealed YbRh$_3$Si$_7$ crystals cannot described by activated behavior between 100 and 300 K. For the optimally-annealed crystal, the resistivity coefficient $A$ is reduced by almost one order of magnitude from $\mu_0 H = 0$ to $\mu_0 H = 9$ T at low temperatures, as shown in Fig. S3(b). This is in line with the fact of Kondo effect being suppressed by applying magnetic fields.

III. NUCLEAR CONTRIBUTION TO THE SPECIFIC HEAT

Below 1 K, the specific heat increases with decreasing temperature due to the nuclear Schottky contribution of Yb element and its isotope, $C_n = \alpha/T^2$, where $\alpha$ is a field dependent constant. At zero field, the energy splitting of the nuclear quadrupole states in the electric field gradient of the atomic environment causes the nuclear contribution to
FIG. S3: (a) $\ln(\Delta \rho)$ vs. $1/T$ plot for as-grown and 200-hour annealed crystals. $\Delta \rho = \rho - \rho_0$. (b) Quadratic temperature dependence of $\rho$ at $\mu_0 H = 0$ and 9 T of the 200-hour annealed crystal. Dashed lines are fits to $\rho = \rho_0 + A T^2$.

FIG. S4: Temperature dependence of the specific heat $C_p/T$ of YbRh$_3$Si$_7$ on a double-logarithmic scale for different fields applied perpendicular to the $c$ axis (empty symbols). After subtraction of the nuclear contribution $C_n$ (solid symbols), $C/T$ remains almost constant below 0.5 K.

IV. CRYSTAL ELECTRIC FIELD PROBED BY INELASTIC NEUTRON SCATTERING

We carried out inelastic neutron scattering measurements on BT-7 for YbRh$_3$Si$_7$ to search for low lying CEF levels, and found that the lowest lying level occurs at about 26 meV, along with significant phonon density-of-states scattering at a slightly higher energy as indicated by its Q dependence. Hence additional measurements were taken on MERLIN at 5, 10, and 100 K on YbRh$_3$Si$_7$, and a nonmagnetic analogue LuRh$_3$Si$_7$ to probe only the phonon scattering, using neutrons with incident energies of 50 meV and 100 meV. Figure S5 shows LuRh$_3$Si$_7$ subtracted scattering from YbRh$_3$Si$_7$, which reveal clear crystal eld levels at 26 meV and 49 meV.
FIG. S5: Q-E map of inelastic neutron scattering of YbRh$_3$Si$_7$, taken on MERLIN at incident energy $E_i = 100$ meV at 5 K and 10 K. Scattering for nonmagnetic LuRh$_3$Si$_7$ was subtracted to probe only the phonon scattering. The CEF levels at 26 meV and 49 meV do not change between 5 K and 10 K.

V. DETAILS OF THE BAND THEORY CALCULATIONS

In the main text, we showed that the DFT+U calculations in the AFM phase predict the $|J = 7/2, m_J = 5/2\rangle$ state as the ground state doublet of YbRh$_3$Si$_7$. For completeness, here we show the results obtained without including the Hubbard $U$ interaction. The projected DOS plot in Fig. S6 shows that the minority $m_L = 2$ orbital (red line) has not fully split off from the other orbitals, unlike in the DFT+U case (Fig. 1(e) in the main text). In this case, we find that the ordered moment is 0.75 $\mu_B$/Yb. The inclusion of Hubbard interaction of $U = 4$ eV on the Yb site further increases the moment to 1.5 $\mu_B$/Yb, a characteristic of the DFT+U framework. Sufficient Hubbard interactions also cause the $m_L = 2$ orbital to split from the other orbitals.

FIG. S6: DOS for the case without Hubbard interactions on Yb. The $m_L = 2$ orbital (red) shows no clear split from the other orbitals. Top(bottom) panels show the minority(majority) spins. The vertical green dotted line represents the Fermi level.

We note that both the DFT and DFT+U values of the ordered moment exceed the experimental value 0.36 $\mu_B$/Yb obtained from the single crystal neutron diffraction. We attribute this disparity to the fact that the DFT-based single-particle framework cannot capture the many-body effects of the Kondo interaction and thus overestimates the bandwidths of Yb $f$-orbitals (and underestimates the effective mass). Additionally, the ordered moments are
FIG. S7: Band structure of YbRh$_3$Si$_7$ in the PM phase with so-called “fat bands” highlighting the contribution from (a) Yb $f$-orbitals and (b) Rh and Si atoms. Thicker sections of the bands represent a larger partial contribution of the respective orbitals. The Fermi surface plots show (c) hole and (d) electron pockets arising from the bands crossing the Fermi level.

Highly sensitive to the position of the Fermi level within the $f$-band. Despite this deficiency, the DFT+U calculations nevertheless provide an adequate qualitative explanation of the magnetic properties, and in particular correctly predict the AFM structure and the nature of the ground state doublet, together with the associated saturated moment (see Fig. 1 and the discussion in the main text).

Further information about the electronic properties in the PM state is obtained from the DFT band structure, shown in Fig. S7 where we have separated the partial contribution of Yb $f$-electrons (Fig. S7(a)) from that of the conduction electrons of Rh and Si (Fig. S7(b)). In this so-called “fat band” representation, the thicker bands denote the larger contribution of the respective atomic orbitals. This figure is to be compared with Fig. 2(c-d) in the main text, the latter computed in the AFM phase using the DFT+U method. The two results are qualitatively the same in that they both show the hybridization between the Yb $f$-electrons and the conduction bands of Rh and Si. The difference is that in the present PM case, all 4 Kramers-degenerate $f$-bands corresponding to $J = 7/2$ states appear close to the Fermi level and hybridize with the conduction electrons, whereas in the AFM case, the DFT+U calculation correctly selects out the $m_j = \pm 5/2$ state which appears at the Fermi level, as described above.

According to our first principles calculations, two bands cross the Fermi level in both the PM and AFM phase. These two bands (the red and navy blue in Figs. S7a-b) form an electron pocket and a hole pocket respectively, which
are shown in Figs. S7(c–d). Because the total number of electrons per unit cell is even, these two pockets are mutually compensated, so that the respective carrier densities $|n_e| = |n_h| \approx 1.6 \times 10^{21}$ cm$^3$ are equal. This rather low carrier density, despite seemingly large Fermi surface pockets seen in Figs. S7(c–d), is due to the fact that the Brillouin zone volume is actually very small, as a consequence of a large real-space unit cell (the rhombohedral primitive cell has volume $V = 375$ Å$^3$). The Luttinger sum rule is indeed satisfied and the two Fermi pockets contain about half of an electron/hole carrier each per unit cell, however given the large unit cell, the corresponding carrier density is low.

**VI. LOW-TEMPERATURE TORQUE DATA**

Figure S8 shows the torque data measured at 0.35 K, which exhibit the same features as the 2 K data shown in Fig. 3(f). Arrows indicate field sweep direction during the measurements.

![Figure S8: Torque at $T = 0.35$ K as a function of magnetic field $H||c$.](image)

**VII. SCHEMATIC VIEW OF THE CAFM PHASE**

The CAFM phase as shown in Fig. 4 in the main text is more clearly illustrated in Fig. S9.

![Figure S9: Magnetic spin configurations in the CAFM phase (left) and net magnetic moment ($M_{001}$) along the hexagonal c axis.](image)
VIII. ANGULAR DEPENDENT MAGNETORESISTANCE

For angular dependent magnetoresistance measurements, the crystal was attached onto a sapphire chip with GE varnish and rotated from the $c$ axis towards the $ab$ plane, as shown in Fig. S10. A Hall probe was used to calibrate the angle where the error bar was about a couple of degrees. The MM transition at $\mu_0 H_1$ moves up in field, from 6.7 T for $H \parallel c$ to 10 T for $H \parallel ab$, and $\mu_0 H_2$ moves above the available field range of 35 T for $\theta \sim 62$ degrees.

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FIG. S10: Angular dependent magnetoresistance of YbRh$_3$Si$_7$ measured at $T = 1.47$ K.