

Magnetic phase diagram of YbRh₂Si₂: The influence of hyperfine interactions

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We report the determination of the magnetic phase diagram of the heavy fermion metal YbRh₂Si₂ in magnetic fields up to 70 mT applied perpendicular to the crystallographic *c* axis. By a combination of heat capacity, magnetocaloric, and magnetoresistance measurements, we map two antiferromagnetic phases: the electronic AFM1 below 70 mK and electronuclear AFM2 below 1.5 mK. The measurements extend into the microkelvin regime to explore the quantum phase transitions in this system. We demonstrate how the hyperfine interaction significantly modifies the phase diagram and the putative field-tuned quantum critical point. The determination of the rich magnetic properties of YbRh₂Si₂ is essential to understanding the interplay of the two magnetic orders and superconductivity in this compound.

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The heavy fermion metal YbRh₂Si₂ has been extensively studied for many years as an example of a system that can be tuned toward quantum criticality, by chemical pressure or by magnetic field [1–7]. Often, superconductivity emerges in heavy fermion metals when they are tuned to the quantum critical point (QCP), the effect of pressure on multiple Ce-based compounds being well established [8]. Following the first Yb-based superconducting heavy fermion β -YbAlB₄ [9], more recently the discovery of superconductivity in YbRh₂Si₂ [10–12] makes it only the second example of an Yb-based superconductor. In a material where superconductivity emerges out of magnetically ordered states, like YbRh₂Si₂, establishing the magnetic phase diagram is essential.

Here, we report a determination of the phase diagram extending into the μ K regime (Fig. 1). It features two antiferromagnetic phases: electronic AFM1 [1] and electronuclear AFM2 [13], each terminating at a field-tuned quantum phase transition. Previously, the critical field of AFM1 had only been mapped down to 20 mK [1]. We study tetragonal single crystals of YbRh₂Si₂ with natural isotopic composition, with magnetic field applied in the *ab* plane. The measurements down to ultralow temperatures required novel calorimetry and electrical transport experimental techniques [12,14]. Both the observed backturn of the critical field of AFM1 and the stability of AFM2 are distinct manifestations of the hyperfine interaction of Yb.

YbRh₂Si₂ features strong anisotropy of the Fermi surface [7,15–19] and electronic magnetism [1]. The AFM1 phase, suppressed by in-plane field of 57 mT or an order of magnitude larger field along the *c* axis [1], features only very small ordered moments, $\mu_e \approx 0.002 \mu_B$ [20], developing out of

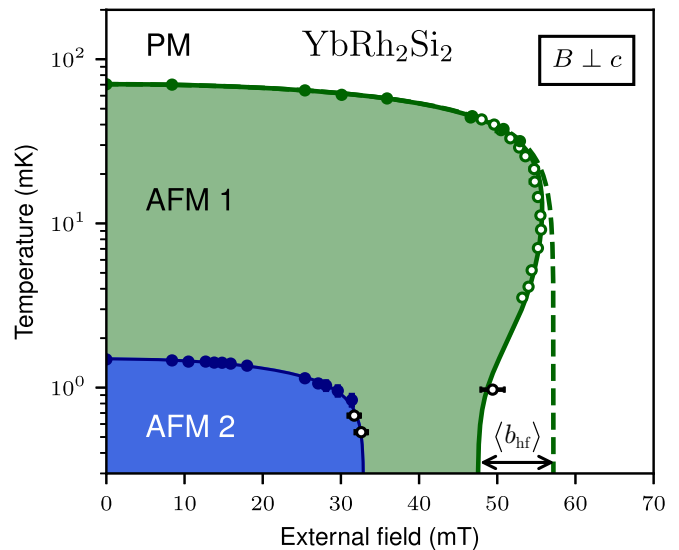


FIG. 1. Phase diagram of the two antiferromagnetic orders in YbRh₂Si₂. The critical field $B_N(T)$ of the AFM1 phase (solid green line) shows a nonmonotonic temperature dependence. We understand this $T \rightarrow 0$ backturn from the asymptote B_N^* (dashed green line) as an effect of the hyperfine field $\langle b_{hf} \rangle$, which adds to the external field according to Eq. (1). The critical field $B_A(T)$ of the electronuclear AFM2 phase shows a regular behavior. The heat capacity peaks are shown by filled circles, magnetocaloric sweep signature by open black circles, and magnetoresistance by open green circles. Calorimetry and transport studies were done on two different samples.

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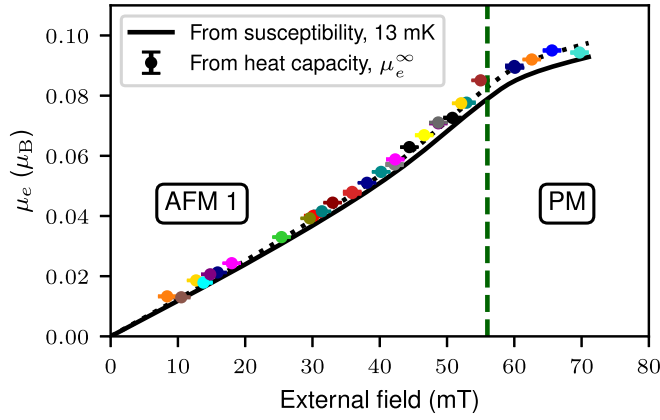


FIG. 2. Growth of the polarized electronic magnetic moment μ_e with in-plane magnetic field at 13 mK, calculated as the integral of magnetic susceptibility [33], and determined from the heat capacity using the *hyperfine Schottky model* [34]. Scaling the susceptibility data by a factor of 1.05 (dotted line) removes a small systematic discrepancy between the two methods.

partially Kondo-screened Yb local moments $1.4 \mu_B$ [1], where μ_B is the Bohr magneton. Ordering of the small moments along the magnetically hard c axis has been proposed [21], but the nature of this order is not understood, and neither is the putative quantum criticality proposed for the suppression of this order with magnetic field. Current theories include the *local quantum criticality* [22–25] (see also [4,5,26–28]) and theories invoking strong coupling of fermions and spin fluctuations into critical quasiparticles [29–32].

A remarkable property of the AFM1 phase is the strong paramagnetic response to in-plane field B_{ext} , manifested by a rapid growth of a static electronic moment μ_e . Figure 2 shows excellent agreement of $\mu_e(B_{\text{ext}})$, inferred from the nuclear heat capacity by a method discussed in detail later in this Letter, with the integral of the ac magnetic susceptibility χ at 13 mK reported in Ref. [33]. This agreement identifies the $4f$ electronic orbital of Yb as the origin of the dominant magnetic effects in YbRh_2Si_2 . The measurements give a single Yb ion susceptibility $\chi_{\text{ion}} \equiv \mu_e/B_{\text{ext}} \approx 1.2 \mu_B/\text{T}$ (at 13 mK and zero field), equivalent to the dimensionless susceptibility $\chi \approx 0.18$. Two properties of the $\mu_e(B_{\text{ext}})$ curve, the inflection at 50 mT and change of slope at 57 mT reflect two features in $\chi(B_{\text{ext}})$ labeled “ B_1 ” and “ B_2 ” in Ref. [33]; the latter feature has been associated with the critical field of AFM1. Thus, established $\mu_e(B_{\text{ext}})$ is the key input for our analysis of the influence of the hyperfine interaction on the AFM1/PM phase boundary.

Recently, we identified the phase, here denoted as AFM2, as an electronuclear spin density wave, the transition to which occurs as a direct result of the hyperfine interaction [13]. The staggered magnetic moments were found to lie in the ab plane and to be continuously suppressed from $\mu_e \sim 0.1 \mu_B$ to zero by in-plane magnetic field. Reference [13] also introduces the use of the heat capacity of nuclear spins as a probe of the electronic moments hyperfine-coupled to them, a method we advance here.

Our primary experimental technique is calorimetry [14], modified from Ref. [13] to allow adiabatic measurements to

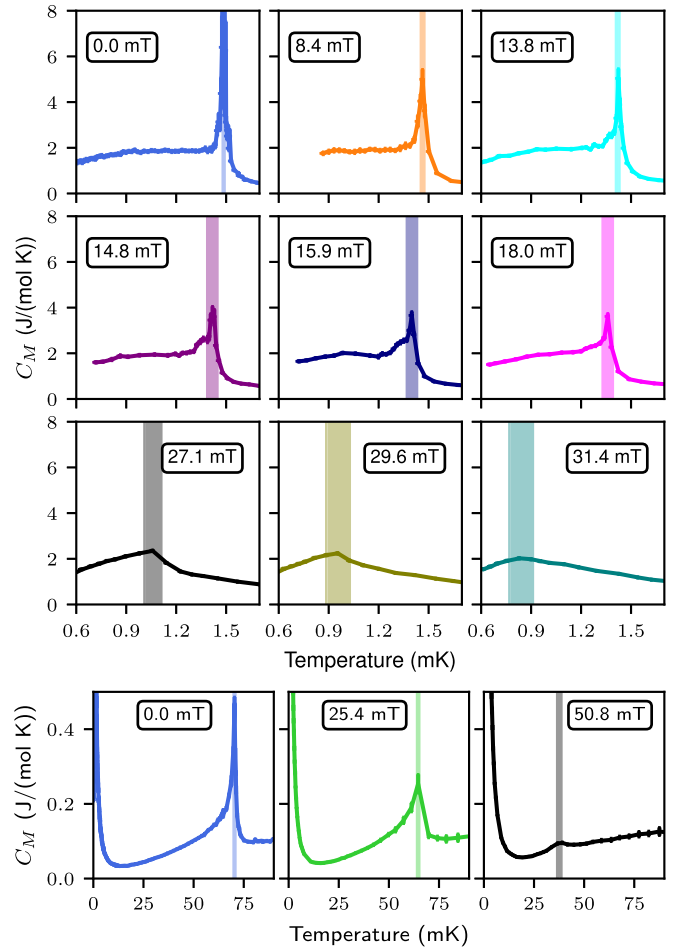


FIG. 3. Evolution of the T_A (top) and T_N (bottom) transition peaks in the molar heat capacity C_M with magnetic field. Estimated transition temperature and its uncertainty are marked by vertical bands. Weak effects of measurement preparation conditions on the position and shape of the T_A transition signature are discussed in the Supplemental Material [34].

higher fields. The heat capacity peaks at T_A (onset of AFM2) and T_N (onset of AFM1) (Fig. 3) give extremely sharp signatures of the transitions in low external field B_{ext} . In both cases, the peaks are suppressed in magnitude with increasing external field. The cusp at T_A in high fields closely resembles the predictions of the mean field theory [13]. The sharpness in zero field may be related to the concurrent transition in the superconducting state [12].

When $\partial T_{A/N}/\partial B_{\text{ext}}$ is high, we must use field sweep methods to identify the phase transitions: the magnetocaloric sweeps (Fig. 4) and magnetoresistance (Fig. 5). The quasiadiabatic magnetocaloric field sweeps [14] allow us to pinpoint the AFM1/PM and AFM2/AFM1 transitions at temperatures below 1 mK, thus close to the quantum phase transition end points. The observed magnetocaloric effect arises from the entropy of Yb nuclear spins, hyperfine-coupled to μ_e , while the electronic entropy is negligible (for detailed discussion see the Supplemental Material [34]).

Crossing the critical field of the AFM1 phase [see Fig. 4(a)], the change of slope of the nearly adiabatic

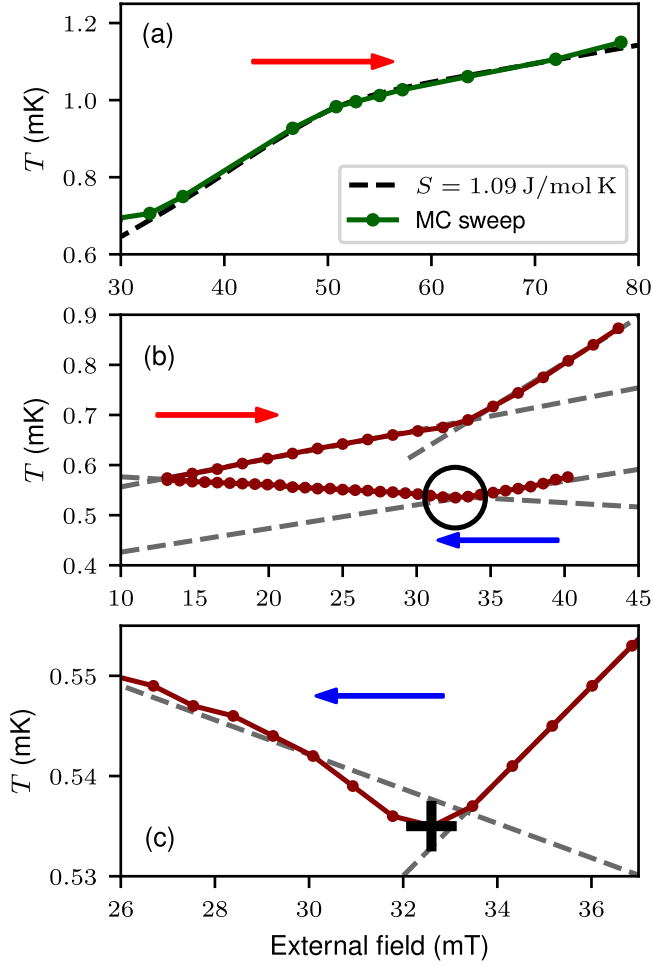


FIG. 4. Examples of quasiadiabatic magnetocaloric sweeps. Direction of the field change indicated by the arrows. (a) Magnetization sweep from AFM1 to PM well described by an isentropic curve (dashed line) derived from the hyperfine Schottky model with $\mu_e(B_{\text{ext}})$ dependence shown in Fig. 2 [34]. This sweep was fast enough for the nonadiabaticity to be negligible. (b) (De)magnetization sweeps across the AFM1/AFM2 boundary with linear fits above and below the transition shown with dashed lines. The negative slope of the demagnetization sweep inside AFM2 is a consequence of parasitic heat deposition into the sample and can be compensated as shown in the Supplemental Material [34]. (c) Detail of the demagnetization sweep in panel (b) highlighted by the circle. A small dip is observed at the AFM1/AFM2 phase transition (in both directions) and here marked by the cross. These features are used as transition points in Fig. 1.

magnetocaloric sweeps is observed, in excellent agreement with the hyperfine Schottky model [34]. This behavior is the response of the nuclear spins to the drop in magnetic susceptibility across this phase boundary [33]. Such a strong nuclear effect masks any magnetocaloric signatures that may arise directly from the suppression of the electronic AFM1 order [35–39].

The AFM1/AFM2 transition is also marked by a change of slope, [see Figs. 4(b) and 4(c)]. The weaker magnetocaloric effect in the AFM2 phase is a consequence of staggered nuclear polarization of the spin-density wave that considerably

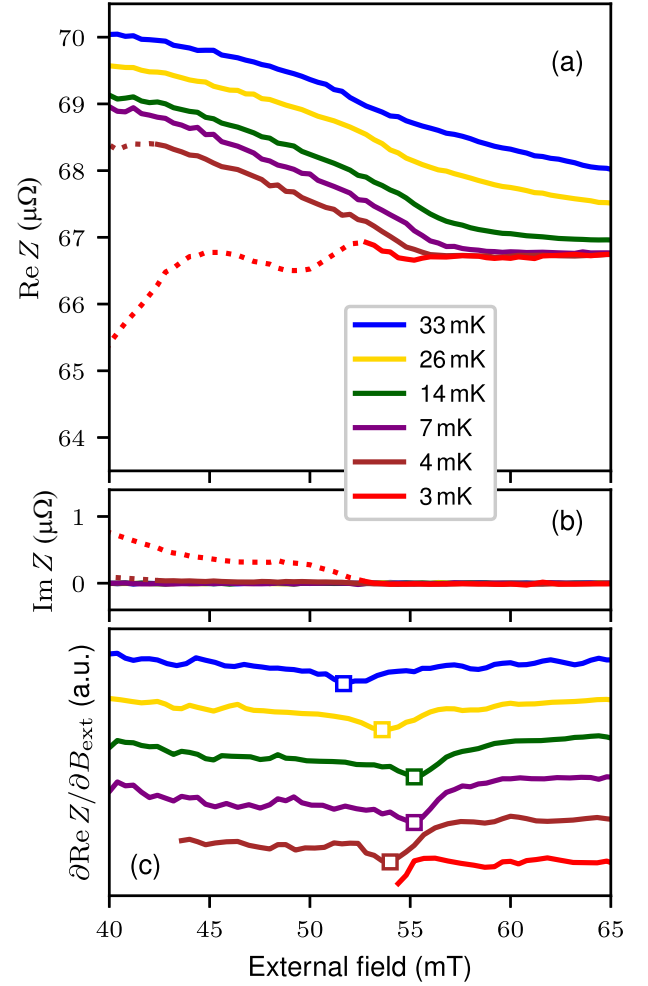


FIG. 5. Measurements of magnetoresistance (a) identify the AFM1/PM phase boundary by a minimum in $\partial \text{Re } Z / \partial B_{\text{ext}}$ (c). (b) At the lowest temperature, this procedure is interrupted by the onset of superconductivity, revealed by the appearance of $\text{Im } Z$ associated with kinetic inductance [12]. On these grounds, the data where $\text{Im } Z > 0.05 \mu\Omega$ shown with dotted lines, were excluded from the analysis. The superconductivity in YbRh₂Si₂ is heterogeneous and does not always manifest as $\text{Re } Z = 0$ [12].

bounds the nuclear entropy [13]. The observed dip [Fig. 4(c)] may be associated with the critical behavior of the electronuclear AFM2 order parameter and/or fluctuations near the transition [35,36].

The AFM1/PM boundary was also determined between 4 and 45 mK from magnetoresistance measurements (Fig. 5), exploiting our SQUID (Superconducting quantum interference device)-based measurements of the complex sample impedance Z with $10 \text{ n}\Omega$ resolution [12]. We identify a minimum in $\partial \text{Re } Z / \partial B_{\text{ext}}$ as the signature of the phase transition. This is justified by the coincidence of this magnetoresistance signature with the heat capacity peaks above 32 mK (see Fig. 1). The magnetoresistance method requires the sample to be in the normal state, so it is interrupted at 3 mK by the onset of superconductivity (see Fig. 5).

Both field sweep methods exhibit no discernible jumps or hysteresis (see Figs. 4 and 5 and Fig. S5 in the Supplemen-

tal Material [34]) across the phase boundaries, suggestive of second-order AFM1/PM and AFM1/AFM2 transitions. This is consistent with the conclusions of earlier work [13,33,40].

The AFM2 dome $B_A(T)$ (Fig. 1) is well described by a cubic polynomial fit to $B_A^2(T)$ (see Eq. (S12) in the Supplemental Material [34]). This allows us to experimentally determine a critical field of 33 mT in the $T = 0$ limit. The data are in good agreement with the line of second-order transitions predicted by the mean field model [13].

The AFM1/PM phase boundary above 20 mK can also be described by a phenomenological ansatz for $B_N^2(T)$ of the same form. However, at lower temperatures, we observe a clear “backturn,” i.e., decrease of the critical field as temperature is lowered, which is fully accounted for by the hyperfine interaction, with the relevant hyperfine field saturating at around 10 mT at the lowest temperatures. Consequently, the external critical field at $T = 0$ is 47 mT, rather than 57 mT conventionally extrapolated from high-temperature measurements [1].

We now explain in detail the procedure to calculate the backturn of the AFM1/PM phase boundary, in terms of known system properties, that is in full agreement with observations (see Fig. 1). The dominant hyperfine interaction in YbRh_2Si_2 involves the $4f$ electron orbitals of Yb [7,41–44] and arises in the Yb isotopes with nonzero nuclear spin: ^{171}Yb ($I = 1/2$) and ^{173}Yb ($I = 5/2$) with natural abundances of 0.14 and 0.16, respectively. The hyperfine constant of Yb is particularly strong, $A_{\text{hf}} = 102 \pm 3 \text{ T}/\mu_B$ [45–48]. In YbRh_2Si_2 , it combines with the particularly strong dependence of $4f$ moment on in-plane field $\mu_e(B_{\text{ext}})$ with dramatic effects.

The hyperfine interaction can be expressed as follows: $U_{\text{hf}} = A_{\text{hf}} \boldsymbol{\mu}_e \cdot \boldsymbol{\mu}_n = -\mathbf{B}_{\text{hf}} \cdot \boldsymbol{\mu}_n = -\mathbf{b}_{\text{hf}} \cdot \boldsymbol{\mu}_e$. Here, $\mathbf{B}_{\text{hf}} = -A_{\text{hf}} \boldsymbol{\mu}_e$ is the hyperfine field exerted by the magnetic moment $\boldsymbol{\mu}_e$ of the electron on the nuclear magnetic moment $\boldsymbol{\mu}_n$; the reciprocal field $\mathbf{b}_{\text{hf}} = -A_{\text{hf}} \boldsymbol{\mu}_n$ is felt by the electron. B_{hf} dominates the nuclear energy level splitting ($B_{\text{hf}} \gg B_{\text{ext}}$) for ^{171}Yb ($I = 1/2$), while in the case of ^{173}Yb ($I = 5/2$), the known quadrupolar interaction [13] (see also Eq. (S3) in the Supplemental Material [34]), must also be taken into account. The temperature-dependent nuclear polarization is determined self-consistently with $\mu_e(B_{\text{ext}})$, taking into account b_{hf} . We note that inside AFM1, the staggered magnetization is insignificant and the hyperfine fields are essentially collinear with the external field.

Figure 6 shows an estimate of $b_{\text{hf}}(T)$ for both isotopes at the critical field of AFM1, using the electronic moment $\mu_e = 0.085 \mu_B$ measured at this phase boundary at 13 mK (see Fig. 2). We find that the backturn is compensated if we add the average $\langle b_{\text{hf}} \rangle$ over the Yb isotopes to B_N . Thus, we propose that the growth of paramagnetic moments μ_e and the suppression of AFM1 order is driven by the *effective field*

$$B^* = B_{\text{ext}} + \langle b_{\text{hf}} \rangle. \quad (1)$$

In terms of this effective field, the AFM1/PM phase boundary $B_N^*(T) = B_N(T) + \langle b_{\text{hf}}(T, \mu_e(B_N^*(T))) \rangle$ is well described by a cubic polynomial fit to $B_N^{*2}(T)$ over the full temperature range (see Eq. (S11) in the Supplemental Material [34]). In the temperature-effective field plane this phase boundary approaches the quantum phase transition vertically, with no backturn (see Fig. S1 in the Supplemental Material [34]).

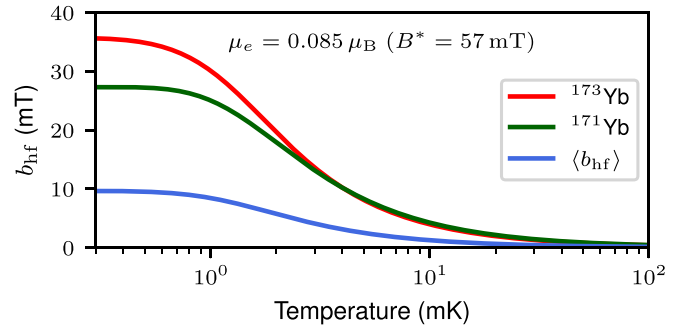


FIG. 6. Hyperfine fields on the ^{171}Yb and ^{173}Yb sites and the spatial average $\langle b_{\text{hf}} \rangle$ over all Yb isotopes as a function of temperature for a constant electronic moment $\mu_e = 0.085 \mu_B$. This value of μ_e corresponds to $B^* = 57 \text{ mT}$, the critical effective field of AFM1 below 13 mK. The quadrupolar splitting of ^{173}Yb in the crystalline electric field is taken into account [13]. Details are provided in the Supplemental Material [34].

The field dependence of electronic moments $\mu_e(B_{\text{ext}})$ at 13 mK, shown in Fig. 2, is inferred from the heat capacity measured between 1.7 and 13 mK, taking into account the gradual increase μ_e on further cooling in a fixed external field driven by $\langle b_{\text{hf}}(T) \rangle$. This improved *hyperfine Schottky model* [34] resolves the discrepancy between the data and the Schottky model with temperature-independent $\mu_e(B_{\text{ext}})$ observed in Ref. [13]. We emphasize that μ_e obtained from this method is in good agreement with the integral of the magnetic susceptibility [33], also plotted in Fig. 2. From these data we derive $\mu_e(B^*)$, assuming it to be temperature independent, and self-consistently calculate $b_{\text{hf}}(B_{\text{ext}}, T)$, feeding into the determination of $B_N^*(T)$ from the measured phase transition signatures [34,49].

Investigating the critical behavior near a QCP [6,50,51] can provide an important insight into the nature of the quantum criticality. We analyze the AFM1/PM transition points as $T = (B_N^*(T = 0) - B_N^*(T))^\varepsilon$ and determine the critical exponent $\varepsilon \approx 0.5$; a similar exercise for the AFM1/AFM2 boundary yields $\varepsilon \approx 0.3$ [34]. These values should be treated with caution because of the limited temperature range and potential effects of disorder.

A detailed study with $B_{\text{ext}} \parallel c$, requiring field-swept measurements in a new higher-field setup, is beyond the scope of this work. A preliminary phase diagram for this field orientation [12] is significantly different from $B_{\text{ext}} \perp c$, with evidence for a PM/AFM1/AFM2 polycritical point, resembling $\text{Yb}(\text{Rh}_{0.82}\text{Co}_{0.18})_2\text{Si}_2$ [21]. The relationship between the magnetic structure of the electronuclear order of the AFM2 phase in YbRh_2Si_2 and the eponymous phase in Ref. [21] remains an open question.

Our results strongly motivate future experiments on isotopically enriched samples using the techniques we have developed. In the absence of active nuclear spins, for example, in $^{174}\text{YbRh}_2\text{Si}_2$ [7], we predict the external critical field of the AFM1 phase to coincide with the $B_N^*(T)$ line, simplifying the determination of the exponent ε , and the AFM2 phase to be absent. On the other hand, we estimate enhanced backturn in $^{171}\text{YbRh}_2\text{Si}_2$ and $^{173}\text{YbRh}_2\text{Si}_2$, where the hyperfine fields saturate in the $T \rightarrow 0$ limit at 27 and 35 mT, respectively (see

Fig. 6). Mean field theory [13] predicts an enhancement in T_A by a factor of 3 in zero magnetic field. Moreover, in these materials the nuclear spins form a regular lattice, in contrast to natural YbRh₂Si₂, which is likely to have significant consequences on the AFM2 phase.

The theoretical framework for treating the impact of hyperfine interactions on quantum criticality has recently been reported [52]. Experimentally, hyperfine effects have been investigated in the transverse-field Ising ferromagnet LiHoF₄ [53,54], where findings include a shift in quantum critical field and softening of collective electronuclear modes near the QCP. In the compound YbCu_{4.6}Au_{0.4}, it was hypothesized that the quantum critical fluctuations do not originate from purely electronic states but from entangled electronuclear states, due to the low Kondo and RKKY (Ruderman-Kittel-Kasuya-Yosida) energy scales [55]. Diverse influences of hyperfine interactions on superconductivity and antiferroquadrupolar order have also been found in PrOs₄Sb₁₂ [56]. The present results open the door to the study of the influence of hyperfine interactions on putative heavy fermion quantum criticality in YbRh₂Si₂.

In conclusion, we report diverse hyperfine effects in YbRh₂Si₂, which has been referred to as a “canonical heavy fermion” material [10]. One finding is that, despite the locality

of the hyperfine fields b_{hf} experienced by the 4*f* electronic shell on the ¹⁷¹Yb and ¹⁷³Yb sites, the entire electron system responds to the average $\langle b_{\text{hf}} \rangle$, manifesting the strongly correlated nature of the heavy fermion state. The present results demonstrate YbRh₂Si₂ as a unique laboratory of heavy fermion physics, featuring interplay between magnetism and superconductivity, and cooperative electronuclear phenomena, with isotopic composition a key tuning parameter in future work.

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The data that support the findings of this article are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

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