

Evolution of Electron Spin Resonance through a Metallic Quantum Critical Phase Diagram

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In the heavy-fermion metal YbRh_2Si_2 , quantum criticality at a suppressed antiferromagnetic order is governed by the interplay of local magnetic moments and itinerant conduction electrons. We demonstrate how this can be investigated by a new experimental approach that enables the observation of electron spin resonance (ESR) across a broad range of frequencies and fields at very low temperatures. This allowed us to cover a large part of the phase diagram from the paramagnetic Fermi-liquid phase to the phase with antiferromagnetic order and including the quantum-critical regime. Both the ESR g factor and the linewidth present distinct behaviors in these three regimes, providing further insight into the physics across a quantum critical point. Notably, when cooling down at a field directly toward the quantum critical point, both g factor and linewidth continuously decrease. Furthermore, we observe a very good matching of the g -factor behavior upon field tuning and temperature tuning toward the quantum-critical point. We analyze and discuss the results in the context of present theories on ESR in strongly correlated electron systems.

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Heavy-fermion metals such as YbRh_2Si_2 are governed by strong electronic interactions. In particular, YbRh_2Si_2 is a realization of a Kondo lattice where the unfilled f shell of each Yb atom in the crystal lattice results in a local magnetic moment, which interacts with conduction electrons. At low temperatures, this so-called Kondo interaction at the Yb sites becomes coherent, local moments and conduction electrons hybridize, and a new metallic heavy-fermion state appears, governed by mobile electrons of strongly enhanced effective mass. This is a nonmagnetic, paramagnetic ground state as the f -derived local moments are Kondo screened by the conduction electrons. If instead the intersite interaction between the moments is dominant, in many cases an antiferromagnetic (AF) ground state appears. The tuning between both ground state regimes, at zero temperature, is a quantum phase transition, which can lead to distinct and unconventional properties at finite temperatures caused by quantum criticality [1,2].

YbRh_2Si_2 is a prime example of such a quantum-critical heavy-fermion system. When cooled in absence of a magnetic field B , YbRh_2Si_2 orders antiferromagnetically below the very low Néel temperature $T_N = 70$ mK [3]. This magnetic order can be suppressed if a moderate magnetic field B , exceeding the critical field of 60 mT, is applied perpendicular to the c axis of the tetragonal crystal structure. The phase diagram is displayed in Fig. 1(a) where at $T = 0$ the quantum-critical point separates the AF order below $T_N(B)$ from a paramagnetic (PM) metallic state at elevated fields. Both states are extremely heavy Landau Fermi liquids [1], in the following labeled “AFL” and “PFL,” respectively. The Landau Fermi liquid (LFL) is a canonical model for conventional metals where electronic interactions are renormalized according to the quasiparticle concept that interprets the metallic many-body state within a single-electron picture. At finite T , there is a wide range in the phase diagram with unconventional metallic properties caused by the presence of the quantum-critical point. This quantum-critical regime features numerous properties that are at odds with LFL theory, and dubbed non-Fermi liquid (NFL). The phase diagram of YbRh_2Si_2 close to this quantum-critical point has been studied in great detail with magnetic, thermodynamic, and transport measurements, and these have not only established the canonical quantum-critical phase diagram but also an additional temperature scale [4,5] which increases with magnetic field. This T^* is considered a manifestation of the crossover from the

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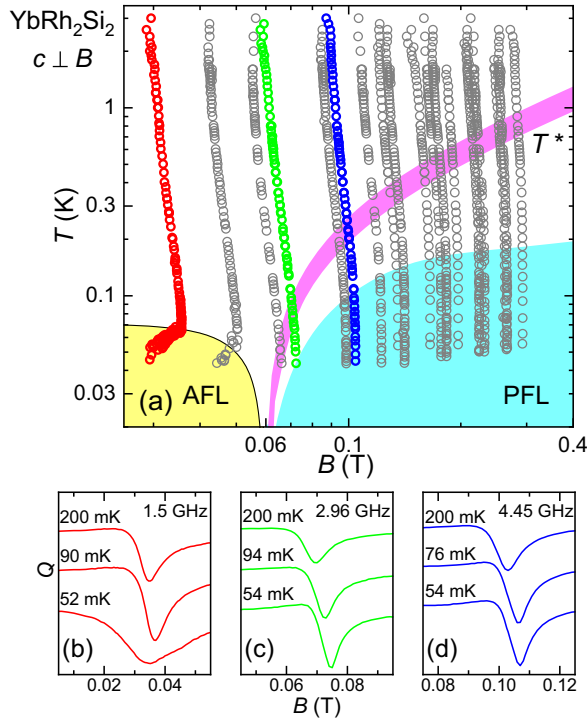


FIG. 1. Probing the phase diagram of YbRh₂Si₂ with ESR. (a) Overall phase diagram with extremely heavy LFL regimes showing AF order (AFL) and metallic paramagnetism (PFL), as well as the T^* line [1]. The circles indicate the ESR resonance field for different microwave frequencies obtained by temperature-dependent ESR measurements; i.e., we probe all relevant regimes. The pronounced evolution of these resonance fields as function of temperature clearly differs for the different regimes, and directly reflects the temperature-dependent g factor. (b)–(d) Exemplary ESR spectra (background-corrected resonator quality factor Q as function of magnetic field B) for three resonance frequencies, with matching colors in (a), for three temperatures each. Key observations upon cooling are (b) broadening and back shift in the AFL phase, (c) monotonous shift and narrowing in the quantum-critical regime, and (d) temperature-independent resonance field in the PFL regime. Spectra for further temperatures are presented in Supplemental Material [26].

low-field state, where the f electrons are of local nature, to a state where they are delocalized and contribute to composite charge carriers. The exact nature, however, is not completely understood [6,7]; alternative pictures were discussed [8], and recent experimental results [9] support the notion that T^* is associated with a partial ferromagnetic (FM) polarization of fluctuating magnetic moments [10]. Moreover, a distinct inelastic scattering process of the heat carriers at T^* [11,12] may be considered a finite-temperature precursor to a violation of the Wiedemann-Franz law at the quantum critical point [11,13,14]. The nature of the AFL phase remains enigmatic: on the one hand a LFL phase forms upon cooling through $T_N(B)$ [4,11,17,18], with effective quasiparticle masses even considerably larger than those in the PFL phase [1], while on the other hand, AF ordering sets in with

staggered moments possibly along the hard crystal-electric-field direction [19,20] as tiny as $2 \times 10^{-3} \mu_B/\text{Yb}$ [21]. Further on, AF “hybrid nuclear, $4f$ -electronic” order [22], or “electro-nuclear,” spatially modulated [23] order onsets at low mK temperatures and is intimately connected with the remarkable formation of a heavy-fermion superconducting phase which is currently considered to be based on a spin triplet pair density wave [16,22–25].

Spectroscopy is the desired experimental approach to address the different energy and temperature scales of YbRh₂Si₂. Unfortunately, the most common spectroscopic techniques such as neutron scattering, scanning tunneling, photoemission, optical spectroscopy, or nuclear magnetic resonance face severe technical limitations for the particular case of YbRh₂Si₂ [6,42–46]. We will show that electron spin resonance (ESR) emerges as a novel spectroscopic tool that is ideally suited for the study of YbRh₂Si₂ close to the quantum-critical point.

ESR is a powerful local probe to address intrinsic magnetic properties. If in the presence of a static magnetic field a transverse microwave magnetic field of frequency ν matches the resonance condition $h\nu_{\text{res}} = g\mu_B B_{\text{res}}$ for the g factor of the material under study (with h Planck’s constant and μ_B Bohr magneton), microwave power is absorbed. The two most important parameters to describe ESR are g , being a property of the resonant magnetic moment, and the ESR linewidth ΔB , which for metals generally corresponds to the transverse spin relaxation rate $1/T_2$ that is governed by the spin dynamics of the surrounding [47,48]. The local moments are screened by Kondo interaction, and commonly the magnetic moments in heavy-fermion metals are not accessible to ESR techniques. Therefore, it came as a great surprise when a pronounced and narrow ESR line was detected in YbRh₂Si₂ [49] and a few other heavy-fermion metals with pronounced FM fluctuations [19,50]. YbRh₂Si₂ and chemically substituted variants now serve as model systems for ESR in Kondo lattices [51–55].

The theoretical understanding developed twofold. A local approach found that, while on-site Kondo screening causes an immeasurably huge linewidth, coherent Kondo scattering in a Kondo-lattice system crucially supports a narrow, measurable ESR signal [55]. This is mainly due to the formation of a collective spin mode of quasilocated f electrons and wideband conduction electrons even in the presence of strong anisotropies in the Kondo interaction [56]. An itinerant approach could qualitatively explain the results by a heavy quasiparticle resonance [57]. Both approaches account for FM correlations, providing an important additional contribution for the narrowing of the resonance lines.

Since conventional ESR spectrometers operate at a fixed microwave frequency and scan the magnetic field, probing the different phases of YbRh₂Si₂ at certain magnetic fields requires different ESR frequencies [51,58,59]. Most of

these experiments were performed at ESR fields of ≈ 200 mT and higher, and all of them were performed at temperatures above 400 mK. This implies that the most interesting region of the phase diagram, at $T < 100$ mK and $B < 100$ mT, was not accessible. Here we overcome previous limitations by introducing a new experimental approach that operates at low-GHz frequencies, being sensitive enough for YbRh_2Si_2 , and compatible with mK temperatures in a dilution refrigerator.

We employ superconducting planar resonators based on a one-dimensional transmission line [60] that can easily be operated at several harmonics. Using meander shaped lines, resonator frequencies as low as 1.5 GHz are implemented on a chip with a size of just a few mm. Resonators of different length enable us to vary the ESR frequency as desired. Using three resonators at $40 \text{ mK} < T < 4 \text{ K}$, we have performed ESR measurements on YbRh_2Si_2 in a wide range of the low-temperature phase diagram, as illustrated in Fig. 1(a). The data acquisition and line-shape analysis are described in Supplemental Material [26]. In the following presentation of our ESR results we start with the low-field AFL region, continue with the PM region at elevated fields, and finally consider the quantum critical regime between AFL and PFL states.

From Fig. 1(a) one can deduce the general temperature dependence of the ESR resonance field for a given resonator frequency within the phase diagram of YbRh_2Si_2 , and this also becomes apparent already from the few exemplary ESR spectra in Figs. 1(b)–1(d). Upon cooling the ESR typically shifts to higher fields and becomes more pronounced, consistent with previous measurements at higher T [49,60]. However, this general trend does not hold at low temperatures. Whereas in the PFL regime the ESR does not change much upon cooling further, the effects on the ESR are dramatic when entering the AFL phase, despite the very weak magnetic moments of the AFL phase [21].

The ESR in and near the AFL phase is illustrated in detail in Fig. 2 for the two resonators with 1.5 and 2.07 GHz: the resonance fields strongly decrease, $g(T)$ correspondingly increase [Figs. 2(b) and 2(d)] and the ESR line becomes much broader than in the PM phase [Fig. 2(c)]. Noteworthy, this evolution of the resonance field is the opposite behavior of the ordered resonance modes in CeRuPO [61] and $\text{Yb}(\text{Rh}_{0.73}\text{Co}_{0.27})_2\text{Si}_2$ [62] which both display FM order along the c axis being the hard crystalline electric field direction. In the region closely above the AFL phase $\Delta B(T)$ tends to saturate or to reach a minimum around 0.3 K while $g(T)$ is continuously decreasing; see Figs. 2(c) and 2(d). The linewidth behavior indicates less effective narrowing due to slowing down of magnetic fluctuations or a reduced effect of FM correlations [50,63]. The temperature and field dependence of the g factor reflect the evolution of the internal field above the ordered state. As detailed in [26] we could describe this by a molecular

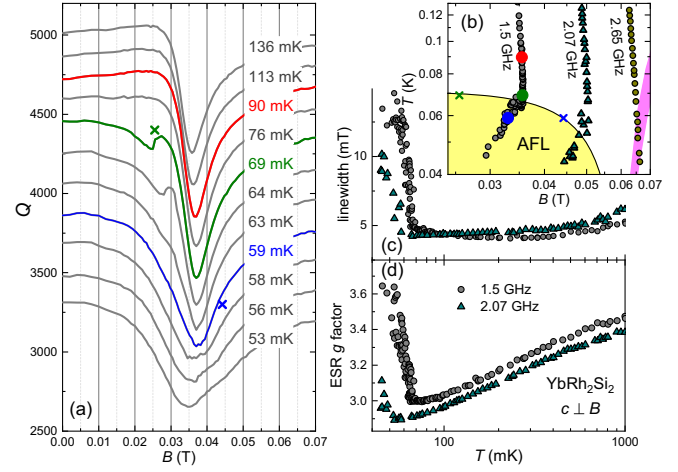


FIG. 2. ESR of YbRh_2Si_2 in the AFL phase and crossing the AF transition. (a) ESR spectra for selected temperatures at 1.5 GHz, including temperature and field ranges in the AFL phase. Q axis refers to 53 mK data, higher temperature data are shifted by 200 each. (b) ESR resonance fields for the three lowest resonator frequencies at low temperatures. Upon cooling, the modes at 1.5 and 2.07 GHz change drastically when entering the AFL phase. Data points at 59, 69, and 90 mK of the 1.5 GHz resonator correspond to colored spectra in (a). The crosses in (a) and (b) correspond to fields of the AFM transition at 59 and 69 mK. (c) ESR linewidth ΔB and (d) g factor increase drastically in the AFL phase for the two lowest frequencies.

field model assuming FM exchange in the plane and AF interaction between the planes. Within this model the resulting g factor in the ordered phase is expected to reach $g_{\text{eff}}^{\text{AF}} \approx 3.72$. This is considerably larger than the observed $g_{\text{exp}}^{\text{AF}} \approx 3.4$, which is probably due to Kondo screening.

ESR spectra in the field region of the AFL phase are presented in Fig. 2(a) for selected temperatures around $T_N(B)$. Their structure is expected to depend on the interplay between internal and external magnetic fields, thus containing information on the magnetically ordered structure as was analyzed for the well-localized magnetic moments in the ordered state of GdRh_2Si_2 [64]. However, the analysis for YbRh_2Si_2 is less clear and the spectra contain structures merely related to the phase boundary $T_N(B)$ as illustrated in Fig. 2(a). The spectra at $T = 69$ and 64 mK, just below $T_N(B = 0)$, exhibit a sharp kink at fields of 25 and 27 mT, respectively. These fields correspond to the critical AF field, as indicated by the green crosses in Figs. 2(a) and 2(b). These kinks can be understood as the result of the transition from a very broad line in the AFL regime at low fields to a narrow line in the PM regime at higher fields. Because at these temperatures the transition occurs at fields well below the resonance field, at the transition the signal of the resonance is already sizable for the broad AFL line, but much weaker for the narrow line of the PM state, resulting in an upward step at the transition from AFL to PM. There seem to be similar features on the

high field side at about 45 mT in the $T \leq 59$ mK spectra which match also quite well with the $T_N(B)$ curve [blue crosses in Figs. 2(a) and 2(b)]. However, these features are much weaker, preventing a strong statement. We note that the simultaneous observation of an AFL and a PM-type ESR in the same spectra does not mean a coexistence of the two phases, since the two signals are observed in distinct field ranges.

Next we turn to the ESR results in the PM region which contains the T^* and PFL regimes. The PFL state of YbRh_2Si_2 is experimentally well established, in particular the T^2 behavior of the dc resistivity served as indicator for the PFL temperature T_{PFL} as a function of field in the phase diagram of Fig. 1(a) [17]. Note, however, that this is not a sharp transition but a crossover between two metallic regimes. As far as the ESR behavior in the PFL regime is concerned, in general one expects a quadratic temperature dependence of the linewidth while the g factor is temperature independent [57]; however, this was never directly observed. In Fig. 3(a) we clearly see the change from a strongly temperature-dependent g factor in the NFL regime above T_{PFL} to a T -independent g factor below. Utilizing the numerous ESR frequencies available to us, we can track the temperature evolution of the T_{PFL} line that fully reproduces the established $T_{\text{PFL}}(B)$ dependence.

From Fig. 3(b) we see that the ESR linewidth exhibits a distinct s-shaped temperature dependence in the PM range.

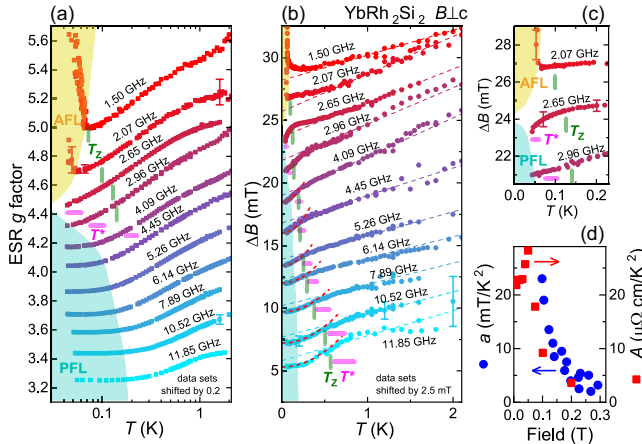


FIG. 3. Temperature dependence of ESR parameters, with focus on paramagnetic regimes. (a) ESR g factor and (b) linewidth ΔB for exemplary resonator frequencies. The transition/crossover temperatures for antiferromagnetic (AFL) / paramagnetic (PFL) regimes are indicated by yellow and cyan areas, respectively; the T^* crossover range is shown by magenta bars, and the temperature T_Z that corresponds to the Zeeman energy ($k_B T_Z \equiv h\nu_{\text{res}} = g\mu_B B_{\text{res}}$) is indicated by vertical lines for magnetic fields close to the critical field. This region is enlarged in panel (c). Straight dashed lines are guides to the eye; parabolic dashed lines represent (aT^2) with field-dependent coefficients a plotted in (d); comparison of a to the quasiparticle scattering cross section $A \propto m^{*2}$ determined from electrical resistivity [17].

Starting with the flattened part in the PFL regime, for $B > 100$ mT ($\nu_{\text{res}} > 4.45$ GHz) the lines significantly broaden above T_{PFL} until $\Delta B(T)$ appears to change around T^* , approaching the known linear dependence for $T > T^*$ [58,59]. Our detailed low-temperature data now allow a much closer look for fields toward the quantum critical field. There the suppression of $\Delta B(T)$ upon cooling becomes pronounced already at the temperature of the Zeeman energy, $k_B T_Z \equiv g\mu_B B_{\text{res}} = h\nu_{\text{res}}$ rather than at T^* , becoming most pronounced near the quantum phase transition as can be well seen for the 2.65 GHz data enlarged in Fig. 3(c). This behavior is in agreement with the critical slowing down seen by THz spectroscopy [65] and is consistent with the expected $T^{3/4}$ behavior for spin-flip scattering of critical quasiparticles [8]. In the regime $B > 100$ mT and $T < T^*$, the linewidth can be well described by $\Delta B(T) = aT^2 + \text{const.}$ as indicated by the dashed lines in Fig. 3(b). The field dependence of the prefactor a resembles a similar behavior of the corresponding factor A in the dc resistivity $\rho(T) = AT^2 + \text{const.}$ [17] observed in the PFL regime [Fig. 3(d)]. Hence, the signatures of PFL are visible in $\Delta B(T)$ up to $T^* > T_{\text{PFL}}$. This could be due to probing the PFL properties by the linewidth being a dynamic quantity in contrast to the static quantities $\rho(T)$ and $g(T)$. An additional narrowing of the lines below T^* can qualitatively be understood by FM correlations stabilizing the resonance [10,50]. Noteworthy, in contrast to the ac susceptibility [5], no maximum occurs in the g factor around T^* [66].

Finally let us consider the quantum-critical regime [Fig. 4(c)], which only now is fully accessible for ESR measurements in temperature and field. In particular the 2.65 GHz data track the T dependence in the NFL regime over 2 orders of magnitude. The continuous evolution in the g factor evidences a logarithmic behavior in a wide temperature range. Such logarithmic dependence early on indicated that a single-ion Kondo scenario was applicable [49]. Later it was shown that the Kondo effect supports the ESR signal by a collective spin mode of Yb spins and conduction electron spins [56]. The same behavior in $g(T)$ is found for all other modes in the NFL regime.

While the field dependence of the ESR parameters, displayed in Figs. 4(a) and 4(b), is rather weak above 500 mK, the behavior changes drastically at lower temperatures. For $T = 125$ mK, the g factor clearly increases with magnetic field, even more pronounced for fields above the T^* scale. At even lower temperatures, $T = 53$ mK, we observe a drastic rise of g for fields above the critical field [26].

Starting from thermodynamic data, Fermi liquid theory allows a renormalization of the quasiparticle Zeeman splitting [8]. As discussed in [26] (Fig. S7), these theoretical estimates yield much larger values and show much stronger field dependencies than the renormalization extracted from the measured g factor, which is a direct spectroscopic experiment. The discrepancies may be a

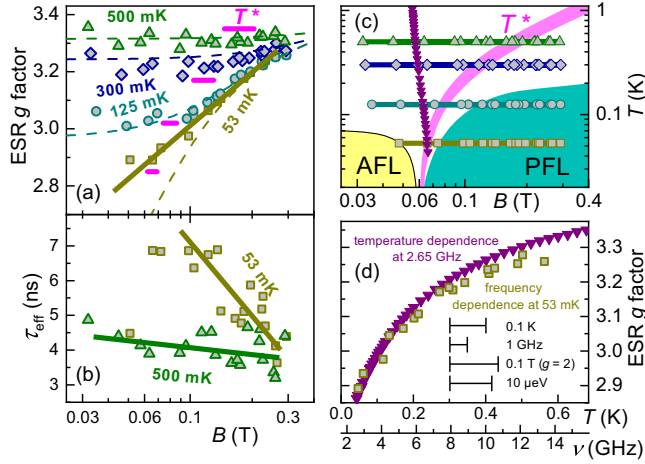


FIG. 4. Magnetic field dependence of ESR parameters in the paramagnetic regimes. (a) g factor and (b) effective ESR relaxation time $\tau_{\text{eff}} = B_{\text{res}}/\nu\Delta B$, for temperatures and resonance fields as indicated in panel (c). Solid lines guide the eye; dashed lines: molecular field model [26]. (c) Phase diagram of YbRh₂Si₂ with horizontal-line connected symbols (resonance fields) referring to data shown in the other frames. The solid triangles denote the resonance fields of the 2.65 GHz mode. (d) Comparison of the g -factor temperature dependence in the quantum-critical regime [horizontal axis: T (K); $\nu = 2.65$ GHz mode] and frequency dependence in the PFL regime [horizontal axis: ν (GHz); $T \approx 53$ mK]. The two horizontal axes as well as additional ranges in the figure indicate the quantitative relation of different relevant energy scales. To achieve matching of the two data sets in (d), the frequency axis is shifted by 1.68 GHz, which corresponds to the ESR frequency for $g = 2$ at the critical field of 60 mT.

result of the dual itinerant-localized character of the f electrons in such strongly correlated f electron systems.

At the lowest temperature of 53 mK, the ESR relaxation time τ_{eff} increases strongly when approaching the quantum-critical field. This is a signature of the coefficient $a(B)$ shown in Fig. 3(d), and reflects the field dependence of the quasiparticle scattering cross section extracted from the dc resistivity [59]. This picture is consistent with the scenario of a fermionic critical slowing down near a magnetic quantum phase transition [65].

The low-temperature g factor increases logarithmically with field well into the PFL regime; this reminds of the similar T dependence in the NFL range. For direct comparison, we make two cuts through the phase diagram in Fig. 4(c): the vertical cut yields $g(T)$ measured at a fixed frequency of 2.65 GHz, i.e., very close to the critical field. The horizontal cut gives the $g(B)$ for the lowest temperature of $T = 53$ mK. Figure 4(d) presents the energy dependence of the g factor for $h\nu$ and $k_B T$. Since the quantum critical point at $T = 0$ and $B = 60$ mT is the conceptual starting point for both dependencies, the origin of the frequency axis has to be shifted by 1.68 GHz. The overlaps of the two datasets is extraordinary and suggests an intrinsic relation

between the frequency/field dependence in the PFL regime and the temperature dependence in the quantum critical regime. We propose to look for the same correspondence in other physical quantities, e.g., the specific heat. The necessary frequency shift of 1.68 GHz corresponds to the ESR frequency at 60 mT for $g = 2$. This could imply that the ESR signal in YbRh₂Si₂ evolves from free-electron-like ($g = 2$) at the QCP to local Yb moments ($g = 3.777$ in this tetragonal crystal environment [67]) at high temperatures and high fields. Clearly, much lower temperatures and precise control of the ESR frequency [68] would be required to check whether the ESR signal really converges to $g = 2$. The g factor approaching $g = 2$ right at the QCP can be regarded the first experimental verification of the theoretically proposed “loss of (composite heavy) quasiparticles at a Kondo-breakdown QCP” [69]. It proves a unique view through ESR into Mott-type quantum criticality, as realized in YbRh₂Si₂.

Tracking the ESR evolution toward the QCP (at the critical field [68] and at lower temperatures) might help to disentangle the role of the static magnetic field as a tuning parameter for both the phase diagram of YbRh₂Si₂ and the ESR frequency. The increase of the linewidth upon entering the AFL phase could be caused not only by a common inhomogeneous broadening effect but also by the notion that resonance-stabilizing FM fluctuations [50] are not relevant in this phase as can be concluded, e.g., from early NMR investigations in the PM region close to the QCP [70]. Overall, our new experimental approach can help to unravel the enigmatic nature of the ordered phase and is able to fully cover the interesting regimes of quantum criticality (NFL) and paramagnetic LFL (PFL). Therefore, it should also be applied to other quantum-critical heavy-fermion materials where an ESR signal was observed, such as β -YbAlB₄ [71], or to other spin systems with novel low-energy phases [72].

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Data availability—There are no publicly available research data or software supporting this manuscript. Requests for further information or data should be sent to the authors.

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