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Conduction electron spin resonance in the α -Yb_{1-x}Fe_xAlB₄ ($0 \leq x \leq 0.50$) and α -LuAlB₄ compounds

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Abstract

β -YbAlB₄ has become one of the most studied heavy fermion systems since its discovery due to its remarkable physical properties. This system is the first reported Yb-based heavy-fermion superconductor (HFS) for which the low- T superconducting state emerges from a non-fermi-liquid (NFL) normal state associated with quantum criticality Nakatsuji *et al* 2008 *Nature* **4** 603. Additionally, it presents a striking and unprecedented electron spin resonance (ESR) signal which behaves as a conduction electron spin resonance (CESR) at high temperatures and acquires features of the Yb³⁺ local moment ESR at low temperatures. The latter, also named *Kondo quasiparticles spin resonance* (KQSR), has been defined as a $4f$ - ce strongly coupled ESR mode that behaves as a local probe of the Kondo quasiparticles in a quantum critical regime, Holanda *et al* 2011 *Phys. Rev. Lett.* **107** 026402.

Interestingly, β -YbAlB₄ possesses a previously known structural variant, namely the α -YbAlB₄, phase which is a paramagnetic Fermi liquid (FL) at low temperatures Macaluso *et al* 2007 *Chem. Mater.* **19** 1918. However, it has been recently suggested that the α -YbAlB₄ phase may be tuned to NFL behavior and/or magnetic ordering as the compound is doped with Fe. Here we report ESR studies on the α -Yb_{1-x}Fe_xAlB₄ ($0 \leq x \leq 0.50$) series as well as on the reference compound α -LuAlB₄. For all measured samples, the observed ESR signal behaves as a CESR in the entire temperature range ($10 \text{ K} \lesssim T \lesssim 300 \text{ K}$) in clear contrast with what has been observed for β -YbAlB₄. This striking result indicates that the proximity to a quantum critical point is crucial to the occurrence of a KQSR signal.

Keywords: conduction electron spin resonance, heavy fermions, non-Fermi-liquid regime, intermetallic compounds

(Some figures may appear in colour only in the online journal)

1. Introduction

The interplay between heavy-fermion (HF) behavior, unconventional superconductivity, and quantum critical non-Fermi-liquid (NFL) regime is one of the most important and

heavily studied problem in condensed matter physics [4, 5]. As such, identifying materials which possess these properties easily tunable in their phase diagrams is a very important task. β -YbAlB₄, the first reported Yb-based heavy-fermion superconductor (HFS), is perfectly suitable in this regard since

its low- T superconducting state emerges from an NFL normal state associated with quantum criticality [1]. Indeed, β -YbAlB₄ has been the focus of intense scientific investigation, both theoretically and experimentally since its discovery [1, 2, 6–11]. In particular, it has been claimed that the unusual local seven-fold symmetry of the Yb site surrounded by the B atoms gives rise to crystalline electric field (CEF) doublets, which generate a strong anisotropic hybridization strength between the Yb 4*f* and the conduction electrons (*ce*). Consequently, a moment-space line defects in the hybridization between the 4*f* and *ce* lead to a quasi-two-dimensional Fermi surface with a non-trivial moment and energy dependence. Such a result has important implications for the quantum critical regime of β -YbAlB₄ [6].

Another important consequence of these interesting CEF effects on the hybridization of the Yb 4*f* electrons with the *ce* in β -YbAlB₄ is the negligible overlap of the CEF wavefunctions of the 4*f* electrons with the in-layer B *p*-bands. This peculiarity has been suggested to be the reason why conduction electron spin resonance (CESR) can be observed in β -YbAlB₄ and β -LuAlB₄ [2, 10, 12] since the *ce* arising from the B-layers would not be subject to the strong spin-orbit coupling associated with the heavy rare-earths ions. Strikingly, for β -YbAlB₄, the CESR signal presents an unprecedented behavior as a function of temperature as it shows the features of CESR at high temperatures and acquires characteristics of the Yb³⁺ local moment ESR at low temperatures. This ESR signal named *Kondo quasiparticles spin resonance* (KQSR) has been previously defined as a 4*f*-*ce* strongly coupled ESR mode that serves as an *in situ* probe of the Kondo *quasiparticles* in a quantum critical regime [2].

Apart from all the above interesting properties, β -YbAlB₄ curiously possesses an iso-stoichiometric structural variant named α -YbAlB₄. β -YbAlB₄ forms in an orthorhombic structure with the *Cmmm* space group, $Z = 4$, and lattice parameters $a = 7.3080(4)$ Å, $b = 9.3150(4)$ Å and $c = 3.4980(4)$ Å, whereas α -YbAlB₄ adopts the orthorhombic structure in the *Pbam* space group with $a = 5.9220(2)$ Å, $b = 11.4730(3)$ Å and $c = 3.5060(5)$ Å [3]. In both polymorphs of YbAlB₄, the Yb and Al atoms are located within the same *ab*-plane, sandwiched between two B-layers. The packing arrangements within the B-layers differ significantly in the two polymorphs, while the Yb/Al are quite similar [3].

Remarkably, α -YbAlB₄ is not a superconductor in contrast to β -YbAlB₄ and it presents a paramagnetic Fermi liquid (FL) behavior at low temperatures. Recently, Kuga *et al* [13] suggested the possibility of the α -YbAlB₄ phase be immersed into an NFL phase as the compound is doped with Fe at the Al-site. This is because either magnetization or specific heat measurements indicate that both α -YbAl_{0.93}Fe_{0.07}B₄ and β -YbAl_{0.94}Fe_{0.06}B₄ exhibit a magnetic order at ~ 9 K, probably of a canted antiferromagnetic type. The increase in the entropy as well as the decrease in the antiferromagnetic Weiss temperature with Fe-substitution in both systems indicates that the induced chemical pressure by Fe-doping is the mechanism responsible for the suppression of the Kondo temperature and the emergence of the Yb 4*f* electrons magnetism [13].

As such, the α -YbAl_{1-x}Fe_xB₄ seems to be a very promising system for an ESR investigation. ESR experiments will reveal whether the system can be tuned from FL behavior (standard CESR signal) towards magnetism (KQSR signal) while Al is continuously substituted by Fe in α -YbAlB₄.

Therefore, the aim of the present report was to confirm the scenario depicted above by means of the ESR experiments of α -YbAl_{1-x}Fe_xB₄ samples with $0 \leq x \leq 0.50$. α -LuAlB₄ with no 4*f* electron contribution has also been investigated as a reference compound.

2. Experimental details

Several single crystals of α -YbAl_{1-x}Fe_xB₄ ($0 \leq x \leq 0.50$) and α -LuAlB₄ compounds were grown from Al-flux as described previously [1, 3]. The crystal structures and phase purity were checked by x-ray powder diffraction. Typical dimensions of the crystals were $\sim 2.0 \times 2.0 \times 0.5$ mm³ and masses of about 10.0 mg. The X-band (9.5 GHz) ESR data were carried out using single crystals. However, gently powdered single crystals were used for the Q-band (34.4 GHz) ESR experiments in order to enhance the signal-to-noise ratio. All the ESR spectra were taken in a Bruker-ELEXSYS 500 spectrometer using appropriated resonators and cool helium gas flow *T*-controller systems. In general, a broad inhomogeneous dysonian-like ESR lineshape was observed in the whole *T*-range for all the investigated samples [14].

3. Results and discussion

It is worth emphasizing here the specific ESR features that would be expected for a KQSR signal in an Yb-based material. For a KQSR signal, the ESR spectra are expected to present a dual behavior (Yb³⁺ local moment-like and conduction electron-like) associated with a coupled ESR response of the conduction electrons and the Yb³⁺ crystal field doublet determined by the particular point symmetry of the Yb-site in the compound. As such, at high temperatures, the KQSR signal is expected to present a *g*-value near to 2 (*g*-value for conduction electrons) and *T*-independent intensity (Pauli-susceptibility). These are features of CESR. The *g*-value is expected to tend to the *g*-value of the particular Yb³⁺ crystal field doublet (which can be anisotropic) as the temperature is lowered and the satellite lines associated with the hyperfine-structure of the Yb isotopes with a non-zero nuclear spin should appear at low-*T*. Eventually, the intensity of the KQSR response may become Curie-like at low-*T*. These *g*-shift (and *g*-anisotropy) and hyperfine lines indicate that the ESR spectra present features of localized moment ESR at low-*T*. The dual behavior in the *T*-dependence of ESR spectra described above characterizes the KQSR signal [2].

Figure 1 presents the room-*T* X-Band ESR spectra for the studied α -YbAl_{1-x}Fe_xB₄ ($0 \leq x \leq 0.50$) single crystals with $H \perp c$. The angular variation of the crystals' orientation with respect to the steady magnetic field was performed using a goniometer properly coupled to the ESR cavity. Within the accuracy of the experiments no spectral anisotropy was observed at any temperature for all our crystals. The strong

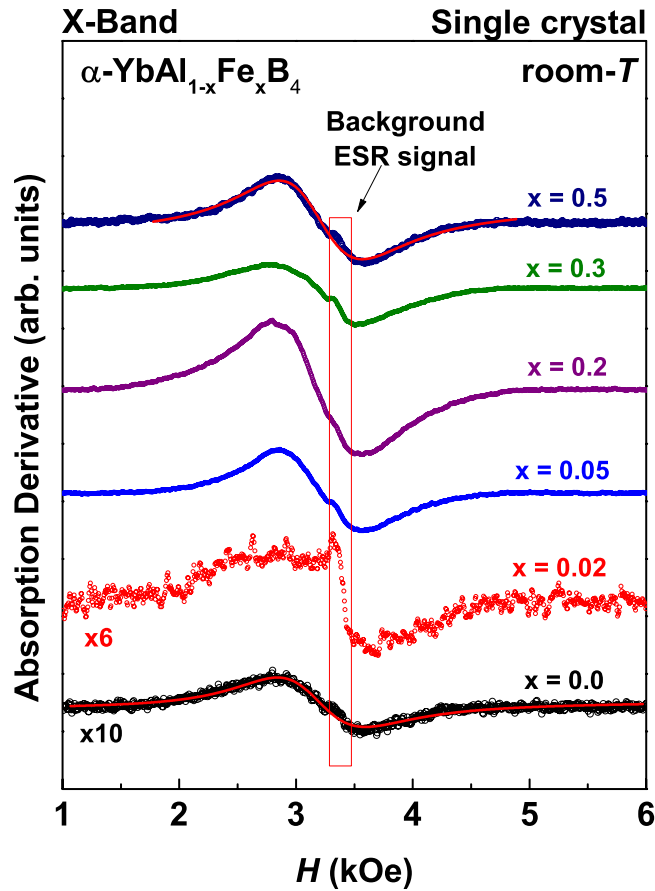


Figure 1. Room- T X-Band ESR spectra for some of our α -YbAl $_{1-x}$ Fe $_x$ B $_4$ ($0 \leq x \leq 0.50$) single crystals with $H \perp c$. The red solid rectangle indicates a background ESR signal contribution. The obtained spectra were analyzed by means of the derivative of the admixture of absorption (χ'') and the dispersion (χ') of the Lorentzian lineshapes according to the Dyson's model for CESR [14]. The following equation shows the expression of the admixture of absorption and dispersion of the Lorentzian lineshapes.
$$\frac{d[(1-\alpha)\chi'' + \alpha\chi']}{dH} = \chi_0 H_0 \gamma^2 T_2^2 \left[\frac{2(1-\alpha)x}{(1+x^2)^2} + \frac{\alpha(1-x^2)}{(1+x^2)^2} \right]$$
 where $x = (H_0 - H) \gamma T_2$, H_0 , and H are the resonance and the applied magnetic fields, respectively, γ is the gyromagnetic ratio ($\gamma = ge/2m_e$), T_2 is the spin-spin relaxation time, α is the admixture of absorption ($\alpha = 0$) and dispersion ($\alpha = 1$), and χ_0 is the paramagnetic contribution from the static susceptibility. The red solid lines are representative fits of the ESR spectra for $x = 0.0$ and 0.05 .

difference between the ESR intensity of the $x = 0$ and $x = 0.05$ crystals may be attributed to either a significant change in the resistivity or to an enhancement of the Pauli magnetic susceptibility of the material.

In figure 2 we show the room- T X-Band ESR spectra for the powdered single crystals of α -LuAlB $_4$ and α -YbAl $_{1-x}$ Fe $_x$ B $_4$ for selected Fe-concentrations. Besides the enormous yet expected increase in the resonance intensity one can observe the distortion of the lineshape and resonance broadening (of the order of the resonance field) towards low fields. This observation is probably caused by the distribution of g -values over a strongly anisotropic Fermi surface [15, 16] and due to the strains produced during the crystal powdering process.

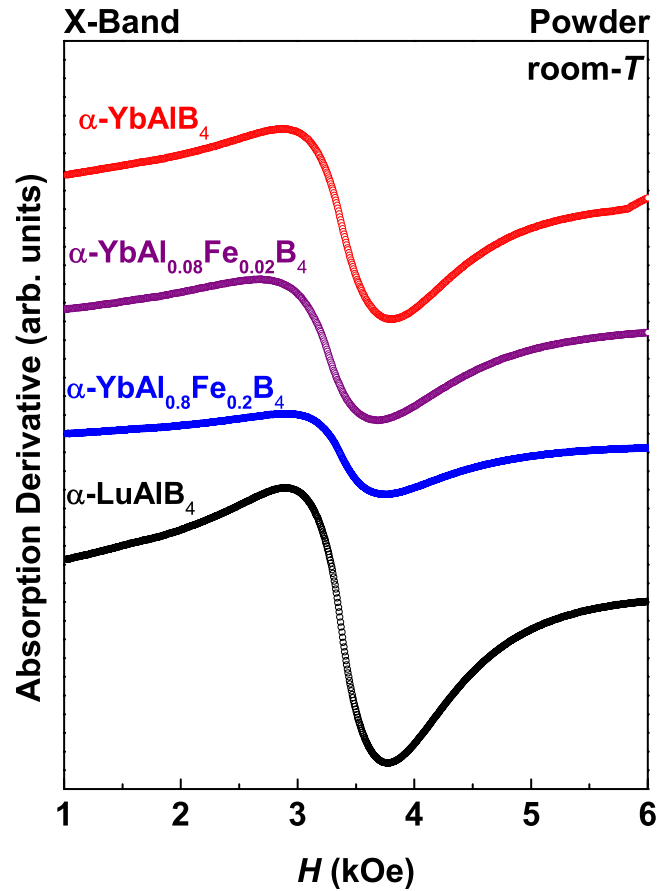


Figure 2. Room- T X-Band powder ESR spectra for α -LuAlB $_4$ and α -YbAl $_{1-x}$ Fe $_x$ B $_4$ ($0 \leq x \leq 0.50$) ($x = 0, 0.02$ and 0.20).

Figure 3 shows, for the representative crystals of figure 1, the T -dependence of the g -value, linewidth, ΔH_{PP} ($\Delta H_{PP} \propto 1/T_2$), and relative integrated ESR intensity I/I_{300K} .

To overcome the poor X-band ESR intensity obtained for the α -YbAlB $_4$ and α -LuAlB $_4$ single crystals and the lineshape distortion observed in the powder samples of figure 2, we displayed the room- T Q-band ESR spectra observed for fine powdered single crystals of these two compounds in figure 4.

Figure 5 presents, for the same powdered crystals of figure 4, the T -dependence of the g -value, the linewidth ΔH_{PP} , and the relative integrated ESR intensity I/I_{300K} , extracted after a Dyson analysis of the ESR spectra [14]. Due to an experimental limitation in our low- T high frequency (Q-band) set up, only data above ~ 100 K could be obtained.

The results shown in figures 3(c) and 5(c) are very important and conclusive. The ESR intensity obtained from the double integral of the ESR spectra is nearly T -independent in the whole studied T -range for all the α -YbAl $_{1-x}$ Fe $_x$ B $_4$ ($0 \leq x \leq 0.50$) and α -LuAlB $_4$ samples. This is a typical CESR behavior since the ce presents a T -independent Pauli magnetic susceptibility. Moreover, it is in contrast to what is expected for the T -dependence of a local moment (LM) ESR intensity, which might present a Curie-law behavior. Besides, we found no resonance anisotropy in any of the studied crystals. Hence, we may conclude that the observed ESR signal is due to the ce ensemble at the Fermi level of the α -YbAl $_{1-x}$ Fe $_x$ B $_4$ ($0 \leq x \leq 0.50$) and α -LuAlB $_4$ absorbed by the microwave.

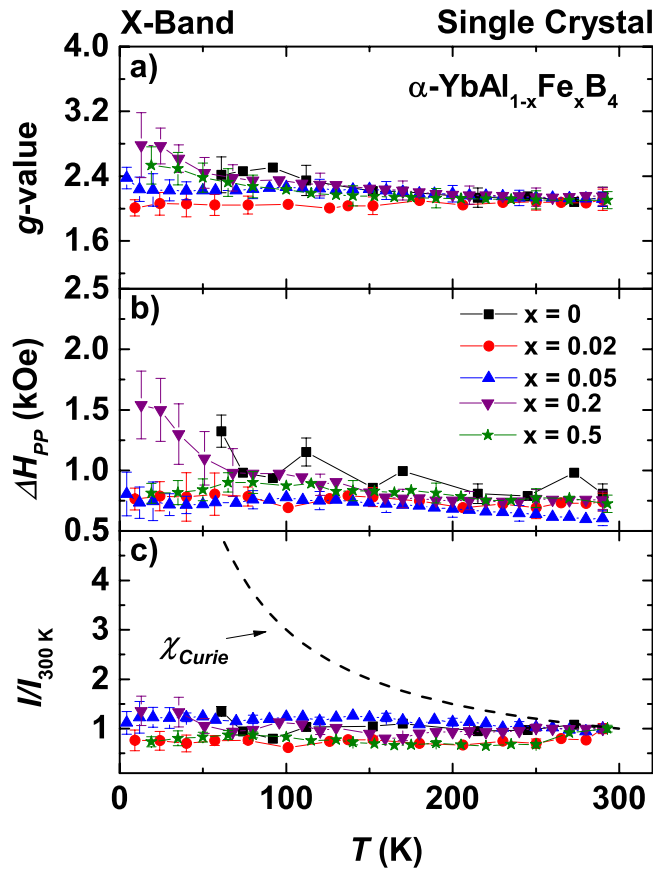


Figure 3. T -dependence of the (a) g -value, (b) linewidth, ΔH_{PP} and, (c) relative integrated ESR intensity, I/I_{300K} , for the spectra of figure 1. The solid lines are guides for the eyes. For comparison, the dashed line in figure 3(c) represents the Curie-law behavior.

Nowadays it is well known that materials such as metals with low ce spin-flip scattering (light metals with small $spin-orbit$ coupling) and/or metals with enhanced Pauli magnetic susceptibility would be prone to presenting an observable CESR [17]. Therefore, a CESR is not expected to be observed due to the strong ce spin-flip scattering (strong $spin-orbit$ coupling) for compounds like β - α -YbAlB₄ and β - α -LuAlB₄ which possess heavy elements as their constituents.

Nonetheless, as for MgB₂ and AlB₂ compounds, where CESR is undoubtedly observed, the above results do suggest that the CESR in the REAlB₄ compounds would be mainly associated with the contribution of the light B and Al constituent elements to the ensemble of the ce at the Fermi level of these materials. The monotonic narrowing of ΔH_{PP} and decrease of the g -value at high- T is certainly accounted for by the scattering of the resonant ce with the lattice phonons via $spin-orbit$ coupling (Elliot mechanism) and by the exchange interaction between ce and the Yb³⁺ magnetic moment (Huber mechanism) [18].

Finally, we should mention that the CESR data obtained for the α -Yb_{1-x}Fe_xAlB₄ ($0 \leq x \leq 0.50$) compounds show no indication for a dual character of the KQSR signal as found for β -YbAlB₄ [2]. Remarkably, in contrast to the β -YbAlB₄, we found no indication of either anisotropy or a g -shift at low- T in the series of α -Yb_{1-x}Fe_xAlB₄ that could be associated with

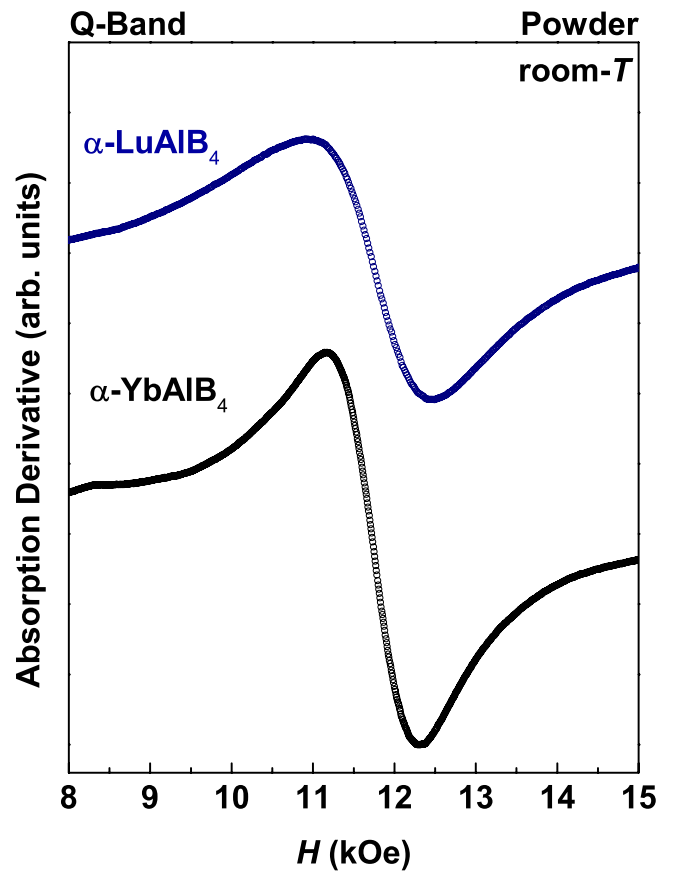


Figure 4. Room- T Q-band ESR spectra of the fine powdered α -YbAlB₄ and the α -LuAlB₄ single crystals.

the ce hybridization of the Yb $4f$ electrons at low- T [10, 19]. Thus, we conclude that the existence of the KQSR signal is restricted to only a very fine tuned critical value of the Yb $4f$ and ce hybridization strength realized in β -YbAlB₄. As such, even though macroscopic measurements may show indications of NFL behavior in the α -Yb_{1-x}Fe_xAlB₄ ($0 \leq x \leq 0.50$), the absence of a KQSR signal in the measured compounds suggests that none of these compounds is tuned to the perfect quantum critical regime such as that achieved in β -YbAlB₄. For low Fe-doped α -Yb_{1-x}Fe_xAlB₄ compounds, the samples are still in the FL regime. Hence, the $4f$ electrons are losing their local moment character and a pure CESR signal is observed. On the other side, for those samples in the high Fe-doping regime, the Yb $4f$ electrons will be too localized and weakly coupled to the ce in such a way that the KQSR mode cannot be realized. Therefore, finding new examples of systems that can exhibit KQSR is proven to be a real non-trivial challenge.

4. Conclusions

We report ESR studies on the α -Yb_{1-x}Fe_xAlB₄ ($0 \leq x \leq 0.50$) series and their reference compound α -LuAlB₄. For all measured samples, the ESR signal behaves as a CESR in the whole temperature range ($10 \text{ K} \lesssim T \lesssim 300 \text{ K}$) in dramatic contrast to the behavior found in β -YbAlB₄. More generally, the ESR results presented here allow us to conclude that B-layered-like compounds, with light constituent elements,

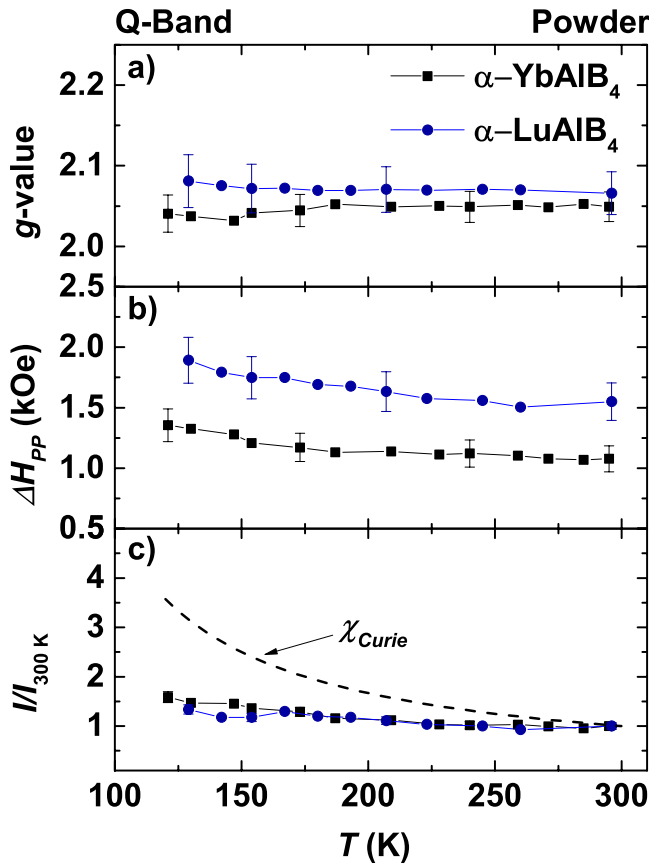


Figure 5. T -dependence of the (a) g -value, (b) linewidth, ΔH_{PP} , and (c) relative integrated ESR intensity, I/I_{300K} , for the spectra in figure 2. The solid lines are a guide for the eye. For comparison, the dashed line in figure 5(c) represents the Curie-law behavior.

may be CESR active depending on their crystal and ce band structures [2, 12]. In summary, the weak T -dependence of the ESR parameters (g -value, ΔH_{PP} , relative intensity) and isotropic behavior of the CESR spectra of the Fe-doped α -YbAlB₄ compounds suggest that a very precise proximity to a quantum critical point is essential to the existence of a KQSR signal.

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