Field dependence of the Eu$^{2+}$ spin relaxation in EuFe$_{2-x}$Co$_x$As$_2$

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Abstract. The layered compound EuFe$_2$As$_2$ is an interesting model system to investigate the effects of well-defined local Eu$^{2+}$ 4f states on the itinerant electronic and magnetic properties of the FeAs layers. To address this subject, we investigated the series EuFe$_{2-x}$Co$_x$As$_2$ (0.1 ≤ x ≤ 0.75) by electron spin resonance (ESR) of Eu$^{2+}$ to probe the spin dynamics of the itinerant subsystem. We relate the results to dc-susceptibility measurements and band structure calculations. As a consequence of the weak coupling between the local and itinerant subsystems, we found that the spin relaxation is well understood in terms of the exchange coupling among the local Eu$^{2+}$ spins. A pronounced field dependence of the Eu$^{2+}$ spin relaxation demonstrates the direct influence of magnetic fluctuations at the Fe$_{2-x}$Co$_x$As$_2$ layers.

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1. Introduction

The discovery of superconductivity in iron-based pnictides [1, 2] sparked a tremendous surge of interest and research activities. This class of materials displays a high structural and chemical flexibility comprising several closely related structure types based on common layered Fe-pnictogen building blocks. Depending on charge-carrier doping, external or chemical pressure, they all show a subtle interplay among structural transitions, antiferromagnetism (AFM) and superconductivity [3]. The relevance of magnetic fluctuations to the stabilization of the superconducting state has been put forward for unconventional superconductors which are found among a wide range of compound families [4–6]. Their role in superconducting iron-pnictides is currently being subjected to intense scrutiny. In this context, the compound EuFe$_2$As$_2$ (with the body-centered tetragonal ThCr$_2$Si$_2$-type structure, where layers of Fe$_2$As$_2$ alternate with Eu layers along the c-axis) was presented as an important model system to study the interplay between well-defined 4f magnetic states and the electronic properties of the iron-arsenide layer [7–9].

EuFe$_2$As$_2$ is an intermetallic compound which undergoes both a spin density wave (SDW)-type AFM phase transition at $T_{N}^{SDW} = 195$ K, associated with the itinerant states of the FeAs subsystem, and a localized AFM ordering taking place at $T_{N}^{Eu} = 22$ K, associated with the Eu 4f local moment subsystem [7, 8]. The magnetic structure of the local moment subsystem consists of Eu$^{2+}$ layers in a ferromagnetic (FM) order, stacked antiferromagnetically along the c-axis [10]. It has been shown that upon substitution on any of the Eu (K substitution) [11], Fe (Co substitution) [12] and As (P substitution) sites [13, 14], the itinerant AFM state is suppressed and the onset of a superconducting phase is observed. Similar behavior is also found when the host compound is subjected to external pressure, which is also claimed to lead to the appearance of reentrant superconductivity [15, 16].

Notwithstanding the large effects observed in the itinerant AFM phase, the energy scale of the local moment interaction, as compared to the energy scale of the itinerant subsystem, has been observed to be only slightly modified in all of these substitutional studies. This suggests a weak coupling between the local 4f electronic states and the electronic and magnetic properties of the FeAs layers. Further indication in this direction is also drawn from the study of the rather weak suppression of the superconducting state by the inclusion of Eu in optimally doped Sr$_{1-x}$Eu$_x$Fe$_{2-x}$Co$_x$As$_2$ [17], from the lack of signatures on the electronic structure of EuFe$_2$As$_2$ when the Eu$^{2+}$ subsystem undergoes the AFM transition [18] and from magnetotransport studies [19].

The nature of the local moment ordering, however, is changed from AFM to FM upon substitution of As by P [13] and possibly also for Fe by Co [20]. In the case of P substitution, it seems clear that the change from AFM to FM order is related to both structural effects and substitutional effects [13]. This is in contrast to the Co substitution, where the change in the electron filling is supposed to be the dominant effect, allowing a more controlled investigation.

In searching for a comprehensive description of the magnetic and electronic properties of the FeAs layers, the electron spin resonance (ESR) of Eu$^{2+}$ has proven to be a suitable tool to provide information on the spin dynamics in EuFe$_2$As$_2$ [21]. In this respect, a proper understanding of the Eu$^{2+}$ spin relaxation is essential, and especially needs to be considered for concentrated Eu$^{2+}$ ions like in EuFe$_{2-x}$Co$_x$As$_2$. In the case of Eu$^{3+}$ ions diluted in a metallic environment, the spin relaxation is determined by the exchange scattering between the local and itinerant spins (the Korringa process) and should be sensitive to the spin dynamics of
both [22]. However, in concentrated magnetic compounds, the electronic spin relaxation is expected to be determined more by the exchange coupling among the local spins [23, 24] which is sensitive to an externally applied magnetic field. Therefore, a systematic investigation of the field dependence of the ESR in EuFe$_{2-x}$Co$_x$As$_2$ and an interpretation in terms of concentrated Eu$^{2+}$ ions provide a unique tool to evaluate the relation between the local moment magnetism and the magnetic and electronic properties of the itinerant subsystem.

To achieve this aim, in addition to dc-susceptibility measurements, we also present the Eu$^{2+}$ ESR at three different magnetic fields, corresponding to resonance frequencies of 1.1 GHz (L-band), 9.4 GHz (X-band) and 34 GHz (Q-band). We shall discuss a significant reinterpretation of the available ESR data [12, 21, 25, 26], which do not include an investigation on the field dependence of the relaxation. The field dependence of the relaxation is a strong piece of evidence that the Eu 4f local moments are coupled with the Fe$_{2-x}$Co$_x$As$_2$ itinerant magnetic fluctuations. Furthermore, we have also performed band structure calculations to guide our discussion on the nature of the Eu$^{2+}$ spin relaxation.

The local moment relaxation is sensitive to the exchange coupling between the local spins by virtue of the so-called bottleneck effect [22]. It occurs when the conduction electron–lattice scattering rate ($1/T_{ceL}$) is smaller than, or comparable to, the conduction electron–magnetic ion ($1/T_{ceI}$) scattering rate. In this situation, after being scattered by the magnetic center (the Korringa process), the conduction electrons, instead of dissipating energy to the lattice, give this energy back to the magnetic ion system (the Overhauser process). As a consequence, one may probe a suppressed Korringa process [22, 27, 28], since the slow $1/T_{ceL}$ will modulate the Korringa rate. However, it may also occur that the relaxation simply does not reflect the exchange scattering between the local moments and the conduction electrons, being determined primarily by the exchange coupling between the Eu$^{2+}$ ions [23, 24].

Even in the latter scenario the properties of the Fe$_{2-x}$Co$_x$As$_2$ itinerant subsystem can be deduced from the local moment relaxation, since the coupling among local spins is realized through the Ruderman–Kittel–Kasuya–Yosida (RKKY) indirect exchange interaction. Hence, while mediating the exchange coupling of the local subsystem, the itinerant subsystem manifests its properties in the local moment relaxation. This interplay qualifies the Eu$^{2+}$ ESR as a suitable probe for the spin dynamics taking place in the Fe$_{2-x}$Co$_x$As$_2$ layers.

2. Methods

Details of the synthesis of polycrystalline samples of EuFe$_{2-x}$Co$_x$As$_2$ (0.1 $\leq x \leq$ 0.75) are given in [20]. The actual content of Co was determined by energy dispersive x-ray scattering (EDX) and is very close to the nominal values. The physical properties of the samples used in this experiment are presented in [20]. The ESR measurements were carried out using a Bruker Elexsys 500 spectrometer for L-band (1.1 GHz), X-band (9.4 GHz) and Q-band (34 GHz) frequencies in the temperature intervals 30 $\leq T \leq$ 120 K for the L-band and 4.2 $\leq T \leq$ 300 K for both X-band and Q-band measurements.

The samples used in the ESR measurements were sieved to a fine powder to achieve grain size homogeneity. In the entire temperature interval, the spectrum consisted of an asymmetric exchange narrowed single broad line. The ESR parameters were obtained by fitting the observed spectra according to the formula given in [29, 30], which also takes into account the effect of a counter resonance in broad ESR lines. The parameters included in the fitting are the ESR linewidth $\Delta H$, the resonance field $H_{res}$, the spectrum amplitude and the lineshape parameter.
\( \alpha = D/A \), which takes into account the dispersion to absorption ratio of the microwave radiation when it probes a metallic surface \((D = 0 \text{ in insulators})\). In the entire temperature interval, the best fitting was always given by this procedure, although we have also tried to search for contributions given by unresolved crystal field (CF) effects and other magnetic anisotropy effects that could be present in the powder spectra. The dc-susceptibility of the samples was measured using a commercial MPMS SQUID dc magnetometer (Quantum Design).

Scalar-relativistic density functional (DFT) electronic structure calculations were performed using the full-potential FPLO code \([31]\), version fplo9.01-35. For the exchange-correlation potential, within the local density approximation (LDA) the parametrization of Perdew–Wang \([32]\) was chosen. To obtain precise band structure information, the calculations were carried out on a well-converged mesh of 4096 \(k\)-points (16 \(\times\) 16 \(\times\) 16 mesh, 405 points in the irreducible wedge of the Brillouin zone). The partial Fe substitution with Co was modeled within the virtual crystal approximation (VCA) as implemented according to \([33]\). Since the structural changes upon partial Co substitution are small in the investigated concentration range, the experimental structural data for the undoped tetragonal \(\text{EuFe}_2\text{As}_2\) compound (space group \(I4/mmm\), \(a = 3.916\ \text{Å}, \ c = 12.052\ \text{Å}, \ z_{As} = 0.3625\) \([34]\) were used throughout the calculations. The strong Coulomb repulsion for the Eu 4f states was treated within the LSDA + \(U\) approximation with \(U = 8\ eV\) as a typical value for Eu. The resulting density of states (DOS) is essentially unchanged for a variation of \(U\) within the physically relevant range.

### 3. Results and discussion

In figure 1(a), we present the temperature \((T)\) versus Co content \((x)\) phase diagram for the series \(\text{EuFe}_{2-x}\text{Co}_x\text{As}_2\), mostly based on our previous investigation \([20]\). The general properties of our ESR investigation for the \(\text{EuFe}_{1.7}\text{Co}_{0.3}\text{As}_2\) sample are shown in figure 1(b) (representative ESR spectra) and in figure 1(c) (ESR intensity \(\chi_{\text{ESR}}\), \(g\)-values). The phase diagram shows that the SDW order is fully suppressed for \(x = 0.2\). This is also the only sample for which we could confirm a transition to a superconducting phase (see the \(T_{sc}\) symbol in figure 1(a)). Furthermore, our magnetization measurements suggest that, for \(x \geq 0.2\), the Eu local moment ordering changes from AFM to FM. These results are quite similar to those obtained by the substitution of As by P \([13, 14]\), where superconductivity is found only in a narrow region of the phase diagram and the nature of the local order also changes on substitution. This similarity suggests that lattice structural effects, rather than the effects of electronic doping, play a fundamental role in tuning the phase diagram in these substitutional studies \([35]\). However, more detailed investigations are required since other works addressing the substitution of Fe by Co reported that a transition to a superconducting state is realized in a much broader interval of Co substitution \([12, 36]\).

Direct inspection of the ESR spectra in figure 1(b) reveals that the change in frequency corresponds to a change in the magnetic fields applied to the sample. We point out that according to the resonance condition \(\hbar \omega = g \mu_B H_{\text{res}}\) the typical fields, for \(g = 2.0\), are roughly 340 Oe for \(L\)-band, 3400 Oe for \(X\)-band and 12 000 Oe for \(Q\)-band. In addition, the \(L\)-band measurements are meaningful only in a relatively narrow temperature interval \((30 \leq T \leq 120\ \text{K})\), since beyond this interval the broadening of the line makes the fitting process highly questionable. Furthermore, we will only discuss the \(L\)-band ESR linewidth \((\Delta H)\).

\(\chi_{\text{ESR}}\) (figure 1(c)) is given by the double integration of the observed spectra. For \(T \geq 40\ \text{K}\) no differences between the \(X\) - and \(Q\)-band results can be resolved. In this temperature region,
**Figure 1.** General characteristics of the investigated samples. (a) Temperature ($T$) versus Co content phase diagram for the EuFe$_{2-x}$Co$_x$As$_2$ series. Arrows indicate the samples measured by ESR. (b) Survey of representative ESR spectra for $x = 0.3$ taken at $L$-band (1.1 GHz), $X$-band (9.4 GHz) and $Q$-band (34 GHz). Magnifying factors are listed on the left side of the corresponding spectrum lines. (c) ESR intensity ($\chi_{\text{ESR}}$) and ESR $g$-values measured at $X$- and $Q$-bands for $x = 0.3$. $\chi_{\text{ESR}}$ displays Curie-like behavior (solid line) as expected for a local moment. The inset shows that at lower temperatures the evolution of $\chi_{\text{ESR}}$ is frequency/field dependent. The ESR $g$-values assume a constant value of $g = 2$ (dashed line) for $T \geq 60$ K.

$\chi_{\text{ESR}}$ follows a Curie–Weiss-like behavior, indicated by the solid line in the figure, as expected for well-defined local moments. In the inset, we show the low-temperature data. The peak observed in the $X$-band $\chi_{\text{ESR}}$ marks the transition temperature of the local moment magnetic order ($T_{\text{EuN}}^\text{Eu}$). This peak is not observed in the $Q$-band $\chi_{\text{ESR}}$ (i.e. at higher fields), suggesting that the nature of the local moment magnetic ordering changes from AFM to FM. However, it indicates that some field needs to be applied to fully stabilize the FM order in the region $x \geq 0.2$ shown in the phase diagram.

The $g$-values (figure 1(c)) are obtained from the resonance condition. For $T \geq 60$ K, they assume a constant value of $g = 2$ that is close to the Eu$^{2+}$ ionic value of $g = 1.993$ found in insulators. At lower temperatures, the $g$-values increase due to the onset of the internal fields associated with the magnetic ordering of the local moments.

In metals, in the absence of a bottleneck effect, it is expected that the resonance position of a given paramagnetic ion is shifted with respect to its value in insulators [22]. This $g$-shift
(Δg = g_{exp} - g_{ms}) is given by

\[ Δg = \langle η(E_F)J(q = 0)\rangle_{Av} = η(E_F) \langle J(q = 0)\rangle_{Av}, \]

where in equation (1) \( \langle J(q = 0)\rangle_{Av} \) denotes an average over the Fermi surface of the \( q = 0 \) component of the exchange interaction \( J(q) \), between the local moment and the itinerant states, and \( η(E_F) \) is a constant DOS for a given spin direction at the Fermi surface (states eV^{-1} mol^{-1} spin^{-1}). Therefore, one expects the \( g \)-values to reflect the evolution of the Fermi surface and magnetic properties of the \( Fe_{2-x}Co_xAs_2 \) layers, both supposed to be induced by Co substitution. In contrast, the \( g \)-values show nearly the same behavior for all the measured samples, indicating that they seem to reflect only the properties of the local moment. This is a feature expected to be found in systems in a strong bottleneck regime [22, 24].

Whereas the \( g \)-values usually reflect static properties, it is expected that \( ΔH \) should be a direct probe of the dynamical behavior of the electronic spin. Experiments on \( EuFe_2As_2 \) [21, 26] single crystals have revealed two distinct types of behavior of \( ΔH \). On the one hand, for \( T > T_{SDW}^N \), a Korringa-like relaxation was found, meaning that \( ΔH = ΔH_0 + b_K T \), where \( ΔH_0 \) is the residual linewidth (\( ΔH \) at \( T = 0 \)) and \( b_K \) is the Korringa rate, which is determined by the exchange scattering of the local moments (the \( 4f Eu^{2+} \) localized states) by the itinerant electronic states (electronic states of FeAs layers). On the other hand, for \( T < T_{SDW}^N \), \( ΔH \) was shown to display an angular dependence but was found to be nearly temperature independent. This was discussed in terms of a suppressed Korringa relaxation due to the gap opening at \( T_{SDW}^N \).

These ideas were drawn on the basis that in metals, in the absence of a bottleneck, the Korringa rate is given by [22]

\[ b_K = \frac{\pi k_B}{g \mu_B} ((N(E_F)J(k_F, k_F'))^2)_{Av} = \frac{\pi k_B}{g \mu_B} η(E_F)^2 \langle J(q)^2 \rangle_{Av} \]

\[ = \frac{\pi k_B}{g \mu_B} η(E_F)^2 J^2, \]

where \( k_B \) is the Boltzmann constant, \( g \) is the measured \( g \)-value, \( \mu_B \) is the Bohr magneton and \( \langle J(q)^2 \rangle_{Av} \equiv J^2 \) is the average over the Fermi surface of the \( q \)-dependent exchange coupling \( J(q) \). In this picture, the gap opening at \( T_{SDW}^N \) would cause \( η(E_F) \) to vanish (or to decrease significantly), thus changing the relaxation regime. The evolution of \( ΔH \) upon Co substitution was also discussed in these terms, although details were not given [12].

The bottleneck effect does introduce an important modification in equation (2). In the bottleneck regime, the apparent Korringa rate \( b \) is given by [22]

\[ b = \left( \frac{1}{T_{cell}} \right) \left( \frac{1}{1/T_{cell} + 1/T_{cell'}} \right) b_K \]

(3)

In the strong bottleneck regime, the exchange scattering will no longer determine the relaxation (at least not in first order), since \( 1/T_{cell} \propto J^2 \). The expression for \( ΔH \) is usually cast in the following form [22]:

\[ ΔH = ΔH_0 + \frac{χ_e}{χ_{dc}} \frac{1}{T_{cell}}, \]

(4)

where \( χ_e \) is the susceptibility of the itinerant electronic system (usually the Pauli susceptibility), and \( χ_{dc} \) is the measured dc-susceptibility of the local moment. It is worth noting that equation (4) mimics the Korringa relaxation but, in contrast, depends strongly on the concentration of the ESR probe. A similar expression can also be deduced by the general arguments given by
Figure 2. The evolution of the linewidth (ΔH) as a function of T obtained for EuFe$_{1.9}$Co$_{0.1}$As$_2$, which undergoes an SDW transition at $T_{SDW} = 100$ K. At this temperature, in contrast to all other samples, ΔH no longer follows a ‘Korringa-like’ behavior (see text). The inset shows in detail that the g-values, unlike ΔH, are unaffected by this transition at 100 K.

Huber [24]. In the following, we shall explain how equation (3) and in particular equation (4) are more appropriate than equation (2) to address the existing experimental data.

We begin discussing figure 2, which shows the X- and Q-band linewidths and g-values (in the inset) for the EuFe$_{1.9}$Co$_{0.1}$As$_2$ sample, which is the only sample investigated displaying an SDW transition. We confirm a linear increase of ΔH for $T > T_{SDW} = 100$ K and a dome-like behavior of ΔH [26] between 40 $\leq T \leq 100$ K (not present in [21]). The g-values (see the inset) are insensitive to the change in the relaxation regime. Furthermore, the relaxation is slightly slower at Q-band.

In principle, both expressions (equations (3) and (4)) may be used to explain the behavior of ΔH. For $T > T_{SDW}^{N}$, one could consider that the linear increase of ΔH originates from a reduced Korringa process or from the inverse of the high temperature $\chi_{dc}$. For $T < T_{SDW}^{N}$, the dome-like behavior of ΔH finds no explanation in the Korringa picture. It could be that it is a coherence peak, due to the gap opening of the SDW transition or, due to its sample dependence, it could be ascribed to the dome which is observed in the resistivity [20]. The resistivity is reflected through $1/T_{\text{rel}}$, which comprises all of the scattering processes (energy dissipation) of the itinerant system. The insensitivity of the g-values to the changes in the relaxation regime indicates a strong relaxation bottleneck.

For all other measured samples (with $x > 0.1$), the evolution of ΔH, as a function of temperature and frequency/field, is well represented by the data in figure 3, which displays the L-, X- and Q-band linewidths for EuFe$_{1.7}$Co$_{0.3}$As$_2$. For $T \geq 40$ K, ΔH broadens linearly with increasing temperature, whereas in the low-temperature region, with decreasing temperature, the proximity of the magnetic phase transition leads to the observed fast broadening of ΔH. The linear increase was fitted to the expression $\Delta H = \Delta H_0 + bT$ for 80 K $\leq T \leq 120$ K in the case of the L-band data and for 100 K $\leq T \leq 300$ K in the cases of the X- and Q-band data. The dashed line shows an extrapolation for $T \geq 120$ K of the L-band fitting result.
Figure 3. Temperature dependence of the linewidth ($\Delta H$) of EuFe$_{1.7}$Co$_{0.3}$As$_2$ for L-, X- and Q-band frequencies. The data are representative for all measured samples, except the $x = 0.1$ sample (for $x = 0.1$ see figure 2). The solid lines are the best linear fit to the data taken at X- and Q-bands above $T = 100$ K. The dashed line is an extrapolation of the low-temperature fit of the L-band data.

The obtained $b$ parameters are compiled in figure 4(a) as a function of frequency/field and Co substitution. As a function of $x$, the $b$ parameter decreases along the series and is nearly constant for $x > 0.4$. With increasing applied frequency/field the $b$ parameter is clearly reduced. This is most pronounced in the region $x \geq 0.2$ where, instead of the now fully suppressed ordered phase of the itinerant subsystem, one expects strong magnetic fluctuations.

The observed behavior of the $b$ parameter on the frequency, being understood as an effect of the frequency-equivalent magnetic field, can be explained by the dependence of the $b$ parameter on $1/T_{\text{cel}}$. In a magnetic compound, the itinerant subsystem will also dissipate energy through a magnetic process, meaning that $1/T_{\text{cel}}$ is also sensitive to spin–spin scattering. A relatively higher field suppresses the magnetic fluctuations in the system, thus lowering the scattering rate of the conduction electrons by the magnetic fluctuations (spin–spin scattering). Therefore, as observed (figure 4(a)), at higher fields $b$ should be smaller.

The evolution of the $b$ parameter as a function of the Co substitution is a somewhat more evolved subject and a number of distinct effects should be considered. First, we consider that the Co substitution contributes to the DOS at the Fermi level [$\eta(E_F)$] by electronic doping. In the case of a relaxation dominated by the Korringa process, $b$ should be proportional to $\eta(E_F)^2$ (see equation (2)). To further investigate this point, we performed band structure calculations to reveal the change of $\eta(E_F)$ upon Co substitution. It has been shown previously that the effect of Co substitution on the SDW, which is closely related to the Fermi surface, can be well described using the VCA [9]. The result for $0 < x < 0.8$ is presented in figure 4(b). $\eta(E_F)$ shows a small upwards variation for $x$ up to $\approx 0.4$ and then starts decreasing continuously for higher values of $x$. This behavior is nearly opposite to that observed in $b$.

The evolution of the local moment fluctuations, as expressed by the Curie temperature $\theta_C$, along the series is presented in figure 4(c). Due to the effects of exchange narrowing, $\Delta H$ would be narrower for larger fluctuations. Accordingly, $b$ should decrease monotonically as
Figure 4. Evolution as a function of Co substitution of (a) the $b$ parameter, obtained at different frequencies/fields, extracted from a linear fitting of $\Delta H$ as a function of temperature, (b) the calculated total density of states (DOS) and (c) the Curie temperature $\theta_C$ obtained from dc-susceptibility measurements.

Figure 5. Plot of $\Delta H$ as a function of reciprocal susceptibility $1/\chi_{dc}$ ($H = 1$ kOe, $T \geq 100$ K). The solid line is a linear fit of the data, and the obtained coefficient $b_\chi$ is presented as a function of Co substitution in the inset.

$\theta_C$ increases. However, this is not the observed behavior, suggesting that the local moment fluctuations alone cannot explain the evolution of the Eu$^{2+}$ spin relaxation as a function of Co substitution.
In figure 5, we present some information relevant to this discussion. We follow the suggestion of equation (4) and show a plot of $\Delta H$ as a function of $1/\chi_{dc}$. It is clearly seen that for a broad temperature interval, $T \geq 100$ K, $\Delta H$ is proportional to $1/\chi_{dc}$. The slopes of these curves ($b_x$), for the entire set of samples (except the $x = 0.1$ sample), are compiled in the inset of the figure. A good qualitative correlation between the concentration dependence of the $b$ parameter and the results of the slopes $b_x$ is found (see the inset of figure 5). Following equation (4), this slope should be written as $b_x \approx \chi_e(1/T_{\text{col}})$, meaning that it expresses an interplay between the magnetism of the itinerant states in the Fe$_{2-x}$Co$_x$As$_2$ layers ($\chi_e$) and the multiple scattering processes taking place in the system ($1/T_{\text{col}}$).

On a technical level, the above results, when put together, strongly suggest that the Korringa process has a marginal role in the spin relaxation in these systems. This means that the Eu$^{2+}$ spin relaxation is driven by the indirect RKKY coupling among the Eu$^{2+}$ spins, i.e. under the influence of the magnetic fluctuations of the itinerant subsystem.

The field dependence of the relaxation reveals the presence of strong magnetic fluctuations after the total suppression of the AFM order of the itinerant subsystem. For $x > 0.4$, where $b$ is nearly constant as a function of $x$, the field dependence of $b$ is strongly pronounced, suggesting stronger magnetic fluctuations than for $x < 0.4$. Furthermore, the field dependence of the relaxation testifies to the existence of magnetic fluctuations even at relatively high levels of Co-substitution in the Fe$_{2-x}$Co$_x$As$_2$ layers.

As already noted, the suppression of the superconducting state by the pair-breaking effect of the Eu$^{2+}$ moments in optimally doped Sr$_{1-y}$Eu$_y$Fe$_{2-x}$Co$_x$As$_2$ was addressed recently [17] and here we use the data of [17] in the subsequent discussion. Pair breaking by a paramagnetic impurity in a superconductor is described by the Abrikosov–Gorkov expression and is driven by an effective exchange scattering that was demonstrated to be comparable with those determined by ESR [37] (see equation (2)). The required DOS for this calculation was shown to be nearly the same for both SrFe$_2$As$_2$ and EuFe$_2$As$_2$ and amounts to $\eta(E_F) = 0.8$ state (eV f.u.)$^{-1}$ [7]. The estimated exchange scattering is $J = 6.1$ meV, which, by equation (2), would drive a relaxation no faster than 1 Oe K$^{-1}$. This should be taken as an upper bound for the value of $b_K$ if the relaxation were determined by the Korringa process. Therefore, this result provides further evidence for the above-described scenario where the relaxation is driven by the exchange coupling among the local spins [24].

In the case of a more traditional bottleneck effect [22, 27, 28], the magnetic fluctuations could increase the apparent relaxation rate by partially, or completely, opening the bottleneck. However, the Korringa rate $b_K$ could not exceed the value of $b_K = 1$ Oe K$^{-1}$ estimated for a full Korringa process. Although not conclusive, this is another important point in favor of an exchange coupled spins scenario to interpret the bottleneck in EuFe$_{2-x}$Co$_x$As$_2$.

4. Conclusions

Investigating the field dependence of the Eu$^{2+}$ spin resonance in the series EuFe$_{2-x}$Co$_x$As$_2$, we found that the electronic properties of the itinerant subsystem are directly reflected in the ESR relaxation mechanism. We have shown that this is achieved in terms of a so-called relaxation bottleneck of exchange coupled Eu$^{2+}$ spins. In particular, this kind of relaxation mechanism is suggested by the opposing evolutions of the linewidth slope ($b$) and the calculated DOS at the Fermi level along the series.
The clear field dependence of the relaxation indicates the presence of magnetic fluctuations and provides a picture of how the relative strength of the itinerant magnetic fluctuations evolves along the series. Interestingly, this field dependence is well pronounced for $x \geq 0.2$, where the AFM order of the itinerant subsystem is suppressed, indicating that it is a property of the emerging itinerant magnetic fluctuations. Furthermore, the field effect is visible even at $x = 0.75$, which is far away from $x = 0.2$.

The ESR results add further evidence that the presence of the Eu$^{2+}$ spins has nearly no effect on the electronic or magnetic properties of the itinerant subsystem. Therefore, one could expect that our picture of the evolution of the magnetic fluctuations is a general property of the Fe$_{2-x}$Co$_x$As$_2$ layers.

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