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Probing putative orbital differentiation effects via Eu^{2+} spin dynamics in in $\text{Sr}_{1-x}\text{Eu}_x\text{Fe}_2\text{As}_2$

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In this work, we report x-ray powder diffraction, elemental analysis, electrical resistivity, magnetic susceptibility, specific heat and electron spin resonance (ESR) in single crystals of $\text{Sr}_{1-x}\text{Eu}_x\text{Fe}_2\text{As}_2$. We observed a breakdown of the previously reported scaling between the Eu^{2+} Korringa relaxation rate obtained from ESR and the spin density wave temperature evolution for Sr-rich samples. This result suggests a distinct evolution of the orbital differentiation of the Fe 3d bands along the Sr-based series when compared to the Ba counterpart. We argue that this difference is related to a larger splitting between the structural (tetragonal-to-orthorhombic) and the Fe-driven spin density wave transitions induced by Eu-doping in this series. In fact, our results indicate that the two transitions follow an opposite x -Eu dependence for Sr-concentrated samples. Our work shows that $\text{Sr}_{1-x}\text{Eu}_x\text{Fe}_2\text{As}_2$ series and the comparison with their Ba-based counterparts are an exciting platform to be explored towards the understanding of the interplay between orbital differentiation, magnetism and structural distortions in the iron pnictides.

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I. INTRODUCTION

The interplay between magnetic order and superconductivity (SC) in several classes of unconventional superconductors suggests that magnetism can play an important role in the pairing mechanism for these systems [1]. Iron-based superconductors (FeSCs) are excellent materials for the study of this relation due to the possible presence of a spin-density-wave (SDW) antiferromagnetic order at high temperatures, which can be suppressed by chemical substitution and/or external applied pressure, inducing a superconducting state [2–4]. However, until now there is no complete microscopic understanding of the origin of the pairing mechanism in FeSCs.[5]

Another element which may be important to the pairing mechanism in FeSCs is the nematic phase [6, 7]. In particular, recent angle resolved photoemission spectroscopy results argue that the microscopic mechanism of the nematic phase should be the same between the iron AFe_2As_2 ($\text{A} = \text{Ba}, \text{Sr}, \text{Ca}$) pnictides and $\text{Fe}(\text{Se}, \text{Te})$ chalcogenides [8–11]. Therefore, there are pressing open questions, such as the role of nematicity [12, 13], orbital selectivity of the itinerant Fe 3d bands [14–22] and, particularly for the iron pnictides, carrier doping [23] to the appearance of a superconducting ground state.

Regarding doping, there are reports of SC induced by substitutions in all distinct crystallographic sites of the iron-based materials [3, 4, 24–26]. It is important to note that isoelectronic P substitution in the As site [3], along with the appearance of SC in stoichiometric iron pnic-

tides under pressure [27], bring insights to the controversy about the actual role of charge doping in these materials [23]. Co-substituted BaFe_2As_2 is regarded as one of the prime examples of charge doping [3, 4]. However, x-ray absorption near edge structure measurements indicate no observable change in the Fe K edge spectra, hence Co-substitution does not add any electrons to the Fermi surface [28]. Furthermore, nuclear magnetic resonance measurements found the same quadrupolar frequency for the diluted regime of Cu^{2+} and Co^{2+} substituted BaFe_2As_2 samples, which again does not support the expectation that each Cu would deliver two extra 3d electrons into the Fermi surface compared to Co substitution [29, 30].

Another crucial point is the role of orbital differentiation in the iron pnictides. This term is usually associated with the distinct weights of the Fe 3d orbitals (d_{xy} , d_{xz} , d_{yz} , $d_{x^2-y^2}$ and d_{z^2}) at the Fermi surface of these materials. This is a result of the actual structural symmetry of the Fe-site and therefore is also strongly connected to tetragonal-to-orthorhombic transition at T_s , as well as to T_{SDW} . Particularly, extended x-ray absorption fine structure measurements have shown that both applied pressure and chemical substitutions in BaFe_2As_2 are responsible for a shortening of the Fe-As bond length accompanied by a suppression of the SDW magnetic phase [31].

Previous electron spin resonance (ESR) reports show that this decrease in the Fe-As distance $d_{\text{Fe-As}}$ is closely connected to the suppression of the SDW magnetic phase

transition temperature T_{SDW} and the localization of the Fe $3d$ electrons in the FeAs planes [32–34]. The increase of the planar character of the Fe $3d$ orbitals, favoring the occupation of the xy and $x^2 - y^2$ orbitals at the Fermi surface may be a key ingredient in the physics of FeSCs [14]. Finally, recent scanning tunneling spectroscopy measurements in $\text{Ba}_{0.6}\text{K}_{0.4}\text{Fe}_2\text{As}_2$ give interesting insights about the orbital selectivity, as it was found that tunneling on each termination surface of this compound probes superconductivity through selecting distinct Fe $3d$ orbitals.[22]. All the above results hint that SC, orbital differentiation of the itinerant Fe $3d$ bands and the Fe-As bond length are intimately connected for pnictides.

In order to further understand and generalize the role of the orbital differentiation to the physics of the FeAs-based materials, it is instructive to tune the coupling between structural and magnetic orders in distinct members of this family. While for BaFe_2As_2 the tetragonal-to-orthorhombic transition at T_s is either a weakly first order or a second order transition [35], for the counterpart compound SrFe_2As_2 the structural transition is first order [36–39]. As a consequence, the structural and magnetic orders are, presumably, strongly coupled, which weakens the nematic phase [39].

In this work we have performed ESR measurements in $\text{Sr}_{1-x}\text{Eu}_x\text{Fe}_2\text{As}_2$ single crystals. ESR is a microscopic probe which can detect changes in the Fe $3d$ bands at the Fermi level through an analysis of the Eu^{2+} spin dynamics [32, 33]. More specifically, the exchange interaction J between the conduction electrons (ce) and the Eu^{2+} local moments depends on the overlap between the atomic orbitals [32, 33]. For $\text{Sr}_{1-x}\text{Eu}_x\text{Fe}_2\text{As}_2$, the Eu^{2+} local moments are our ESR paramagnetic probes located out of the FeAs plane. Therefore, an increase of the planar xy orbital contribution would decrease the exchange interaction between ce and the Eu^{2+} moments. Conversely, an increase in the yz or xz orbital contributions would increase orbital overlap with the ESR probe. Such exchange interaction will be reflected in the relaxation of our probe, through the spin-flip scattering between the Eu^{2+} local moments and the carriers (Korringa mechanism) [41–43], and in the g -value, extracted from the Eu^{2+} resonance field [41–43]. Our data unveil that the Korringa relaxation rate b , and consequently the exchange J , does not follow T_{SDW} in Sr-rich samples. This result indicates that the orbital differentiation of the Fe $3d$ orbitals is instead connected with T_s . We argue that T_s and T_{SDW} follow an opposite dependence as a function of Eu^{2+} substitution in Sr-rich samples. Our work shows that the $\text{Sr}_{1-x}\text{Eu}_x\text{Fe}_2\text{As}_2$ system may be an interesting playground to understand the role of different interactions in the physics of FeSCs.

II. METHODS

Single crystalline samples of $\text{Sr}_{1-x}\text{Eu}_x\text{Fe}_2\text{As}_2$ were grown by the In-flux method [44, 45]. The $I4/mmm$ crystalline structure was confirmed by x-ray powder diffraction at room temperature using a commercial diffractometer. The synthesized phase and the actual Eu^{2+} content in the samples were determined by elemental analysis using energy dispersive x-ray spectroscopy (EDS). High-temperature X-band ($\nu = 9.34$ GHz) ESR measurements were performed in a commercial spectrometer using appropriated resonators and temperature-controller systems. Both single crystals and powdered samples were used in the ESR experiments to study anisotropic effects and to increase the ESR signal-to-noise ratio, respectively. Nuclear magnetic resonance (NMR) experiments in single crystals of undoped SrFe_2As_2 were carried out using a NMR probe equipped with a goniometer for fine in situ alignment of the crystallographic axes with the external applied magnetic field. The field-swept ^{75}As NMR spectra ($I = 3/2$; $\gamma/2\pi = 7.2919$ MHz/T) were obtained by stepwise summing the Fourier transform of the spin-echo signals. Magnetic susceptibility measurements were performed in SQUID-based commercial magnetometer. Specific heat measurements were done in a small-mass calorimeter system that employs a quasi adiabatic thermal relaxation technique. In plane electrical resistivity was obtained in a commercial low-frequency equipment using the standard four-probe technique and with the current applied in the ab -plane.

III. RESULTS AND DISCUSSION

Figure 1 displays the lattice parameters obtained from Rietveld refinements of the $\text{Sr}_{1-x}\text{Eu}_x\text{Fe}_2\text{As}_2$ x-ray powder diffraction. Both a and c decrease linearly with increasing Eu^{2+} content, indicating a homogeneous substitution of Sr^{2+} by Eu^{2+} , in agreement with Vegard’s law [46]. However, c is more affected (1.6 % total variation) than a (0.4% total variation) with increasing Eu^{2+} substitution. The lattice parameters present similar values as in previous reports [2]. No changes on Fe-As local geometry were detected in these measurements along this series within the experimental uncertainty.

Figure 2 presents the specific heat (c_p), magnetic susceptibility (χ) and resistivity (ρ) as a function of temperature for $\text{Sr}_{1-x}\text{Eu}_x\text{Fe}_2\text{As}_2$. One can clearly see a sharp peak at high temperatures in Fig. 2 (a), which can be associated with both the structural and SDW phase transition. One can see that this anomaly initially increases and becomes sharper for the $1-x = 0.97$ sample and then decreases as a function of Eu^{2+} concentration. This decrease is consistent with the expected behavior of the T_{SDW} since EuFe_2As_2 shows a lower T_{SDW} (≈ 192 K) when compared to SrFe_2As_2 (≈ 205

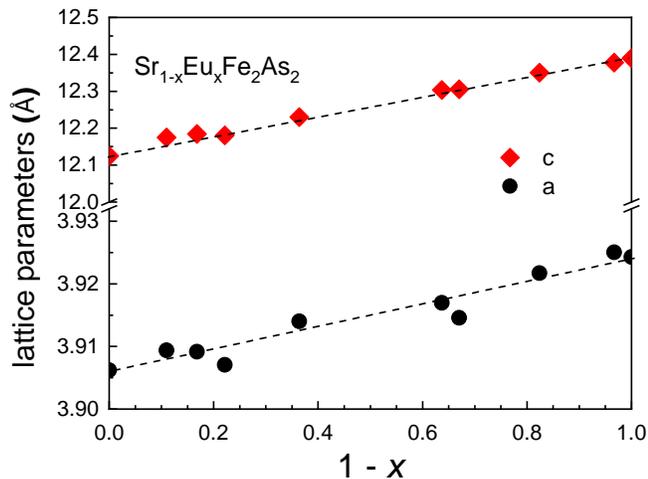


FIG. 1. Lattice parameters a and c for $\text{Sr}_{1-x}\text{Eu}_x\text{Fe}_2\text{As}_2$. The dashed lines are guides to the eye.

K). A small shoulder is visible at intermediate concentrations, which could be related with disorder effects producing a distribution of T_{SDW} s in the crystals. In contrast to substituted BaFe_2As_2 samples, no splitting of T_S and T_{SDW} is observed in specific heat measurements for $\text{Sr}_{1-x}\text{Eu}_x\text{Fe}_2\text{As}_2$. At low temperatures, a rise in c_p/T is observed for Sr^{2+} -rich samples. Such rise is caused by Eu^{2+} - Eu^{2+} interactions, similar to $\text{Ba}_{1-x}\text{Eu}_x\text{Fe}_2\text{As}_2$ [32]. For higher Eu^{2+} concentrations ($1-x \leq 0.36$), antiferromagnetic order of the Eu^{2+} local moments occurs, which generates a low-temperature peak in c_p/T .

The magnetic susceptibility as a function of temperature is displayed in Fig. 2 (b). The magnetic field $H = 300$ Oe was applied parallel to the ab plane. The data for Eu^{2+} -substituted samples, corrected for the core diamagnetism, were fitted to a Curie-Weiss law at high temperatures ($220 \text{ K} \leq T \leq 300 \text{ K}$) (see the top right inset of Fig. 2). From these fits we estimate the Eu^{2+} concentration, assuming each ion carries an effective magnetic moment of $7.94 \mu_B$. All Eu^{2+} concentrations are in agreement with the concentration obtained from EDS measurements. We also extracted the Pauli contribution $\chi_0 = 0.002(3) \text{ emu/mol-Oe}$ for all concentrations. Regarding the magnetic transitions, T_{SDW} can be seen as a kink in χ for all concentrations, as shown in one example in the top left inset of Fig. 2 (b). At low temperatures, one can see a clear monotonic decrease of T_N as a function of Eu^{2+} concentration, which is consistent with specific heat measurements.

The temperature dependence of normalized resistivity (ρ/ρ_{300K}) is illustrated in Fig. 2 (c). At high temperatures, a metallic behavior is observed for all x . A clear kink characterizes T_{SDW} for all samples. The derivative of ρ only shows one peak at high temperatures (left inset) in agreement with the heat capacity data. The estimated transition width from these data for the pure SrFe_2As_2

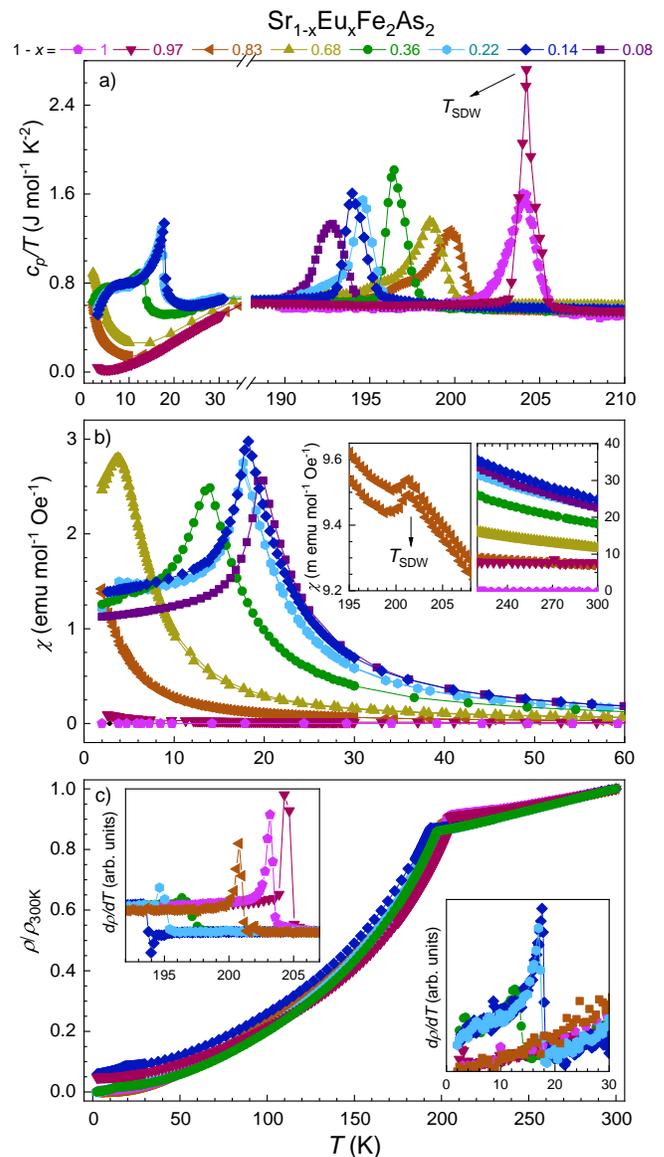


FIG. 2. Temperature dependence of (a) specific heat, (b) magnetic susceptibility and (c) dc resistivity for $\text{Sr}_{1-x}\text{Eu}_x\text{Fe}_2\text{As}_2$. χ measurements were done with applied field $H = 300$ Oe parallel to the ab -plane. The top insets in (b) show the signature of T_{SDW} and the data at high temperatures. The insets in (c) show the derivative of the resistivity near T_{SDW} (left) and T_N (right).

samples is ≈ 2 K, which indicates that if there is a separation between T_s and T_{SDW} in these sample, it should be smaller than 2 K. The right inset of Fig. 2 (c) shows the derivative of the resistivity at low temperatures, in which we obtained a peak that can be associated with T_N . In fact, the transition temperatures extracted from resistivity, specific heat and magnetic susceptibility are consistent with each other.

To gain microscopic insights regarding the possible splitting of T_S and T_{SDW} in $\text{Sr}_{1-x}\text{Eu}_x\text{Fe}_2\text{As}_2$ we have

performed NMR experiments in single crystals of undoped SrFe_2As_2 . Figure 3 shows the ^{75}As NMR signal for an In-grown SrFe_2As_2 single crystal within the 204 K to 200 K temperature range with $H \perp c$. At 204 K, the narrow line at $H \approx 9.35$ T corresponds to the $(-1/2 \leftrightarrow 1/2)$ transition in the tetragonal/paramagnetic phase of SrFe_2As_2 near the structural/SDW transitions. The intensity of the resonance dramatically disappears within a 2 K range as the sample is cooled through the structural/SDW transitions. In addition, no broad line associated with the orthorhombic phase or ordered magnetic domains with strong magnetic interactions emerges in this range, as previously observed for doped BaFe_2As_2 . [58] This result reveals that the presence of even stronger magnetic interactions below T_{SDW} in SrFe_2As_2 are affecting the NMR line associated to the orthorhombic/ordered phase. As a result, the resonance line is wiped out due to quite fast relaxation below T_s . [58]

The presence of even stronger magnetic interactions in the ordered state SrFe_2As_2 when compared to BaFe_2As_2 indicates that most of the available entropy of the structural/SDW transitions rely on the magnetic transition. This would make it harder to track the T_s evolution in the $\text{Sr}_{1-x}\text{Eu}_x\text{Fe}_2\text{As}_2$ series using macroscopic measurements.

Now, to obtain microscopic insights about the orbital differentiation through the Eu^{2+} spin dynamics evolution along the series, we have performed ESR in $\text{Sr}_{1-x}\text{Eu}_x\text{Fe}_2\text{As}_2$ crystals. Fig. 4 shows the summary of our Eu^{2+} X-band ESR. The left panel of Fig. 4 shows the Eu^{2+} ESR spectra at $T = 200$ K for $1 - x = 0.83, 0.68, 0.22$ and 0.14 . The magenta solid lines are the best fits to the Eu^{2+} ESR spectra considering an admixture of absorption and dispersion derivatives. The power absorption derivative (dP/dH) as a function of the applied field H can be expressed as

$$\frac{dP}{dH} \propto (1 - \lambda) \frac{d}{dx} \left(\frac{1}{1 + x^2} \right) + \lambda \frac{d}{dx} \left(\frac{x}{1 + x^2} \right), \quad (1)$$

where the first term is the absorption component, the second term is the dispersion component, λ is the asymmetric parameter of the (Dysonian) line shape, and $x = 2(H - H_r)/\Delta H$, wherein H_r is the Eu^{2+} resonance field and ΔH the Eu^{2+} ESR linewidth. The Dysonian line shape was used to fit the ESR spectra of Figure 4 and extract the Eu^{2+} ESR g -values and linewidth. [42, 43, 47, 48].

The right panel of Fig. 4 shows the temperature evolution of Eu^{2+} ΔH for $200 \text{ K} \leq T \leq 300 \text{ K}$ for different Eu^{2+} concentrations. Towards high temperatures there is a clear linear increase of ΔH , which suggests a Korringa relaxation mechanism [42, 43]. This linear ΔH increase as a function of temperature is observed for all samples. Performing a linear fit of $\Delta H(T)$, $\Delta H = \Delta H_0 +$

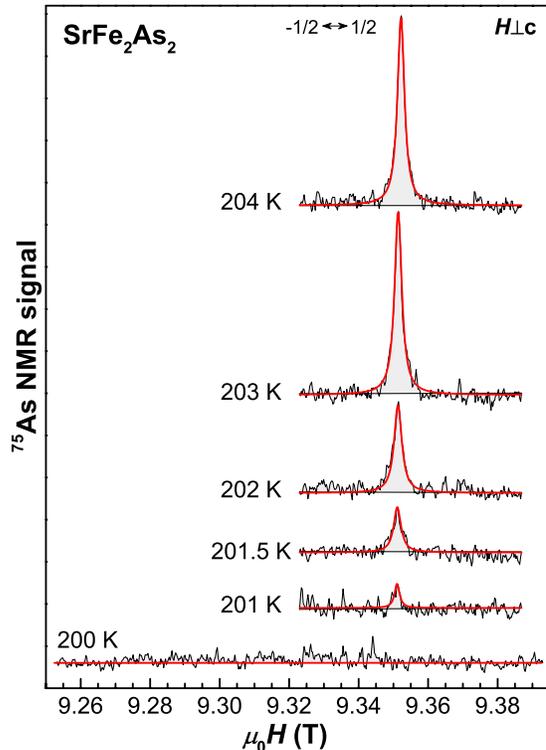


FIG. 3. ^{75}As NMR central transition ($-1/2 \leftrightarrow 1/2$) at $\nu = 68.5878$ MHz for the In-grown SrFe_2As_2 single crystal, within the 204 K to 200 K temperature range with $H \perp c$. The red solid lines are Lorentzian fits to the spectra.

bT , where ΔH_0 is the Eu^{2+} residual linewidth, we obtain the Korringa rate b for all concentrations. For EuFe_2As_2 we obtained $b = 6.6$ Oe/K, which is in good agreement with previous reports [32, 49, 50]. The g -factor of the Eu^{2+} ESR spectra is $g \approx 2$ for all temperatures and concentrations - right inset of Fig. 4.

Figure 5 displays the phase diagram for $\text{Sr}_{1-x}\text{Eu}_x\text{Fe}_2\text{As}_2$ as a function of $1 - x$. Both T_{SDW} and T_N were obtained using the macroscopic measurements detailed above. For comparison, we also present the evolution of the Korringa relaxation rate b as a function of the concentration $1 - x$. We note that T_{SDW} increases and T_N decreases with the decrease of Eu^{2+} concentration. For $1 - x \geq 0.75$ the Eu^{2+} AFM transition is no longer identified above $T = 2$ K. Interestingly the Eu^{2+} AFM ordering in $\text{Sr}_{1-x}\text{Eu}_x\text{Fe}_2\text{As}_2$ has higher T_N than in $\text{Ba}_{1-x}\text{Eu}_x\text{Fe}_2\text{As}_2$ indicating a larger Eu^{2+} - Eu^{2+} magnetic interaction [36–39].

It is worth noting the difference in the trend of the b rate as a function of $1 - x$ in comparison with $\text{Ba}_{1-x}\text{Eu}_x\text{Fe}_2\text{As}_2$ [32, 49, 50]. In the Ba series, the b rate follows the same trend of T_{SDW} as function of Eu-concentration, whereas in $\text{Sr}_{1-x}\text{Eu}_x\text{Fe}_2\text{As}_2$, the b rate

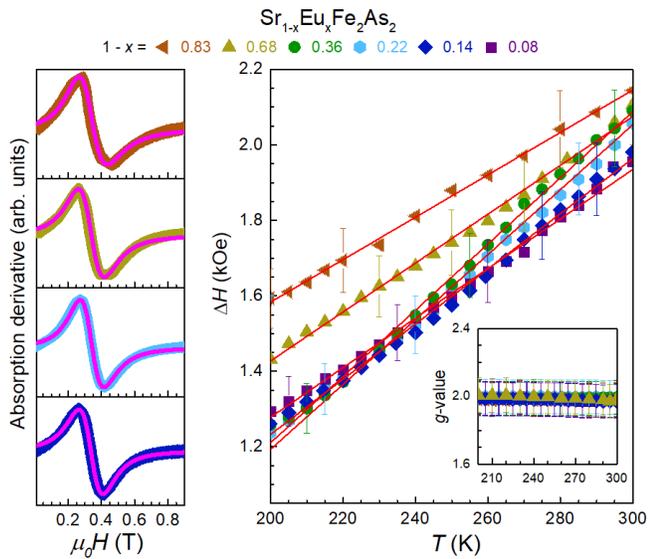


FIG. 4. Left panel: X-band Eu^{2+} ESR spectra at $T = 200\text{K}$ of $\text{Sr}_{1-x}\text{Eu}_x\text{Fe}_2\text{As}_2$ for $1-x = 0.83, 0.68, 0.22$ and 0.14 (no ESR signal was observed for $1-x = 0.97$). The magenta solid lines are fits described into the text. Right panel: Temperature dependence of the linewidth ΔH . The red solid lines are the best linear fits obtained. The right inset shows the temperature dependence of the g -value.

has the same trend of T_{SDW} only at higher concentrations of Eu^{2+} ($1-x \leq 0.5$).

This difference in the Korringa rate b scaling between Sr and Ba compounds can be understood by analysing the exchange interaction J between the Eu^{2+} local moments and the ce in these compounds. Assuming that bottleneck, dynamic, electron-electron correlation, \mathbf{q} -dependence, and multiple bands effects are not present in the interaction between the Eu^{2+} local moments and the Fe $3d$ ce [34], the b rate and the ESR g -shift can be written as [42, 43]

$$b = \frac{\pi k_B}{g\mu_B} J_{fd}^2 \eta^2(E_F), \quad (2)$$

$$\Delta g \equiv g - g_{insulator} = J_{fd} \eta(E_F), \quad (3)$$

where J_{fd} is the effective exchange interaction between the Eu^{2+} local moment and the ce in the absence of ce momentum transfer, $\eta(E_F)$ is the density of states for one spin direction at the Fermi surface, κ_B is the Boltzmann constant, μ_B is the Bohr magneton, and $g_{insulator} = 1.993$ is the Eu^{2+} g -value in an insulator. When Eqs. 1 and 2 are applicable we have the following identity

$$b = \frac{\pi k_B}{g\mu_B} \Delta g^2. \quad (4)$$

Fig. 5 shows that the b rate has a dependence with Eu^{2+} concentration, while the g -value is concentration independent. This is a clear indication that eq. 4 is no longer valid and we should take into consideration the \mathbf{q} -dependence of the exchange interaction, $J_{fd}(\mathbf{q})$ [32, 33, 43]. At the Eu^{2+} site, the g -shift probes the ce polarization ($\mathbf{q} = 0$) and the Korringa rate probes the average over the Fermi surface of the ce momentum transfer ($0 \leq q \leq 2k_F$) [42, 43]. In other words, The Eu^{2+} ESR linewidth Korringa rate, b , depends on the average of the q -dependent effective exchange interaction between the Eu^{2+} local moment and the Fe $3d$ conduction electrons averaged over the whole Fermi surface, $J_{fd}(\mathbf{q})$, which is strongly connected to the Fe $3d$ orbital differentiation and the weight of the Fe $3d$ orbitals (d_{xy} , d_{xz} , d_{yz} , $d_{x^2-y^2}$ and d_{z^2}) at the Fermi surface.

Bottleneck effects are not present because, in the diluted regime, the Korringa rate increases with increasing Eu^{2+} concentration [42]. If dynamic effects were present, the g -value should be T -dependent, which is not the case [51]. Finally, multiple bands effects can be neglected due to the contribution at the Fermi surface being only from Fe $3d$ electrons. The remaining point is the contribution of electron-electron interactions. The estimated Pauli magnetic susceptibility, assuming a free ce gas model, for SrFe_2As_2 ($\chi_{theoretical} \approx 62 \mu\text{emu/mol-Oe}$) is two orders of magnitude smaller compared to the experimental value $\chi_0 = 2(3) \mu\text{emu/mol-Oe}$, which means that electron-electron correlations are important to our ESR analysis. This is consistent with recent inelastic neutron scattering results in BaFe_2As_2 , which also show the importance of electron-electron correlations in iron pnictides [52, 53]. With the \mathbf{q} -dependence and electron-electron interaction assumptions, eqs. 1 and 2 should be rewritten as

$$b = \frac{\pi k_B}{g\mu_B} \langle J_{fd}^2(\mathbf{q}) \rangle \eta^2(E_F) \frac{K(\alpha)}{(1-\alpha)^2}, \quad (5)$$

$$\Delta g = J_{fd}(\mathbf{0}) \frac{\eta(E_F)}{(1-\alpha)}, \quad (6)$$

where $(1-\alpha)^{-1}$ is the Stoner enhancement factor [54, 55] and $K(\alpha)$ is the Korringa exchange enhancement factor [56, 57].

Importantly, the Eu^{2+} ESR g -value is x -independent (Inset of Figure 4). Therefore, we can infer that the density of states for one spin direction at the Fermi surface $\eta(E_F)$ and the Stoner enhancement factor α are nearly x -independent within the experimental error bars (see Eq. 6). Consequently, the q -dependent effective exchange interaction averaged over the whole Fermi surface, $\langle J_{fd}^2(\mathbf{q}) \rangle$ is more affected by doping than its value for $q = 0$, which clearly indicates an evolution of the topology of the Fermi surface with x , since $J_{fd}(\mathbf{q})$ is just the Fourier transform of $J_{fd}(\mathbf{r})$.

Previous studies in $\text{Ba}_{1-x}\text{Eu}_x\text{Fe}_2\text{As}_2$ series have shown a decrease in $\langle J_{fd}^2(\mathbf{q}) \rangle$ as T_{SDW} is suppressed, indicating that the ce bands which overlap with Eu^{2+} $4f$ states are becoming more anisotropic, *i.e.* the d_{xy} orbitals are increasing their role at the Fermi surface. In real space, this is reflected by the increasing of the separation between the Eu^{2+} site and the ce , which characterizes a localization of Fe $3d$ conduction electrons in the FeAs plane. As Ba^{2+} and Eu^{2+} are isoelectronic, there is no introduction of carriers, therefore the evolution in the electronic structures is likely due to subtle changes in the tetragonal crystal structures (and consequently in the Fe-As bonds). Hence, the orbital differentiation and the occupation of the Fe $3d$ orbitals should follow the structural distortion T_s , which is coupled to T_{SDW} in BaFe_2As_2 [32, 33].

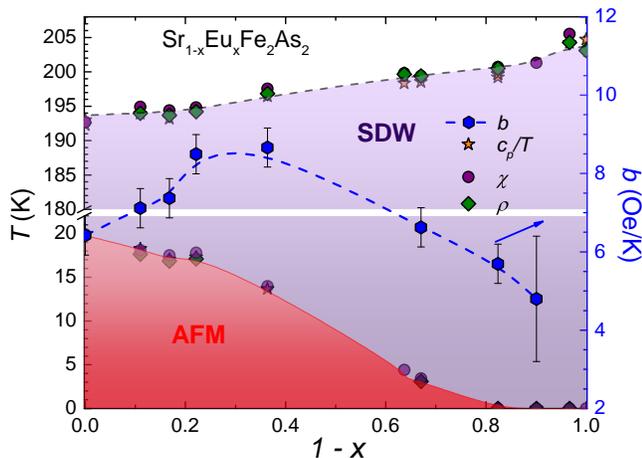


FIG. 5. $\text{Sr}_{1-x}\text{Eu}_x\text{Fe}_2\text{As}_2$ phase diagram and the comparison with the Korringa rate as a function of Eu^{2+} concentration. The solid and dashed lines are guides to the eyes.

In the low Eu^{2+} -concentration regime for $\text{Sr}_{1-x}\text{Eu}_x\text{Fe}_2\text{As}_2$, we have obtained a different scaling between b and T_{SDW} - while the first increases, the second decreases, apart from the possible small initial increase of the both T_s and T_{SDW} for the $1-x = 0.97$ sample. This is a hint that there T_s and T_{SDW} are not tracking each other for Sr rich samples, most likely because of the coexisting magnetically ordered orthorhombic and paramagnetic tetragonal domains are more likely to persist over a larger temperature range above T_{SDW} in the Eu-substituted SrFe_2As_2 compared with their BaFe_2As_2 counterparts [32, 33, 36–39]. This effect was clearly observed in Raman scattering, synchrotron x-ray diffraction, specific heat, resistivity and magnetic susceptibility measurements performed in $\text{Sr}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ [$x = 0.20(3)$] single crystals. [40].

Microscopically, the increase of b reveals an enhancement of $\langle J_{fd}^2(\mathbf{q}) \rangle$ as T_{SDW} is suppressed, indicating a larger contribution of the $d_{xz/yz}$ orbital character. This increase in the axial ($d_{xz/yz}$) orbital contribution is related to an increase in T_s as the SDW phase is sup-

pressed, suggesting a possible initial splitting between T_s and T_{SDW} as they follow opposite directions as a function Eu-concentration in the Sr-rich regime. Presumably, apart from the initial increase of both T_s and T_{SDW} where the anomalies in our data become sharper, T_{SDW} starts to decrease with the larger amount of entropy associated with it, which makes T_s practically undetectable in our macroscopic measurements. However, the Eu- x dependence of T_s can still be tracked by our ESR measurements.

Therefore, in contrast with previous reports [32, 33], we argue that the separation between T_s and T_{SDW} is within a 2 K range for pure SrFe_2As_2 , but they are following an opposite evolution as a function of Eu-doping in the low Eu^{2+} -concentration regime. This occurs most likely due to the strong magnetic fluctuations present in this compound [32, 33, 36–40] which allows the coexistence of orthorhombic and tetragonal domains over a larger temperature range above T_{SDW} and below T_s . In the high Eu^{2+} -concentration region, $1-x \leq 0.5$, the suppression of the SDW phase follows the suppression of the Eu^{2+} ESR Korringa rate, consistent with a localization of the $3d$ electrons in the FeAs plane, similar to the BaFe_2As_2 series which indicates that T_s is accompanying the same trend of T_{SDW} . However, it is worth to emphasize that modeling the microscopic interaction between Fe $3d$ electrons and the Eu^{2+} ESR probes is an open challenge and further experiments such as nuclear magnetic resonance, x-ray absorption fine structure and high resolution x-ray diffraction as a function of temperature in the Eu-doped $(\text{Ba,Sr})\text{Fe}_2\text{As}_2$ could be useful to understand this main difference between the BaFe_2As_2 and SrFe_2As_2 compounds.

V. CONCLUSIONS

In summary, we present a detailed experimental characterization of In-flux grown $\text{Sr}_{1-x}\text{Eu}_x\text{Fe}_2\text{As}_2$ single crystals by x-ray powder diffraction, elemental analysis, specific heat, magnetic susceptibility, electrical resistivity and electron spin resonance (ESR). Our ESR results may indicate a different evolution of the orbital differentiation of the Fe $3d$ bands as the spin density wave is suppressed compared to the Ba series. We suggest that this anomalous evolution is related to an Eu-induced higher splitting between the structural phase transition and the spin-density-wave transition in the dilute regime for $\text{Sr}_{1-x}\text{Eu}_x\text{Fe}_2\text{As}_2$. These results suggest that, indeed, the orbital differentiation of the $3d$ bands of Fe in these materials seems to be governed by subtle structural distortions.

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