Correlated Trends of Coexisting Magnetism and Superconductivity in Optimally Electron-Doped Oxypnictides

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We report on the recovery of the short-range static magnetic order and on the concomitant degradation of the superconducting state in optimally F-doped SmFe1–xRu x AsO0.85F0.15 for 0.1 ≤ x ≤ 0.5. The two reduced order parameters coexist within nanometer-size domains in the FeAs layers and eventually disappear around a common critical threshold x c ≈ 0.6. Superconductivity and magnetism are shown to be closely related to two distinct well-defined local electronic environments of the FeAs layers. The two transition temperatures, controlled by the iso electronic and diamagnetic Ru substitution, scale with the volume fraction of the corresponding environments. This fact indicates that superconductivity is assisted by magnetic fluctuations, which are frozen whenever a short-range static order appears, and totally vanish above the magnetic dilution threshold x c.

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The investigated polycrystalline SmFe1–xRu x AsO0.85F0.15 samples are the same as those of Ref. [8]. From 19F nuclear magnetic resonance the relative fluorine content was found to be constant within Δ ≲ 0.01 in the whole set of samples investigated. To investigate the bulk character of the superconducting state we carried out transverse field (TF)-μSR measurements, where a sample is field cooled (FC) in a magnetic field larger than the lower superconducting critical field Hc1, applied perpendicular to the initial muon-spin direction (H ⊥ Sμ). A flux line lattice is thus generated below Tc and the muon-spin precessions around the local field, Bμ, display a diamagnetic shift Bμ = μ0H(1 + Nμ/2), with χ < 0 and Nμ an effective demagnetization factor. For samples with χ < 0.1 the time evolution of the full TF-μSR asymmetry is fitted to a single SC component as A TF(t) = aTF e−γtτ/2 cos(γBμt), where aTF is the amplitude, γ = 8514μs−1/T is the muon gyromagnetic ratio and τTF the time decay of the precession. This fit holds over the entire T range (not shown) as expected in a bulk superconductor. The shift of Bμ with respect to the applied field is displayed vs temperature in Fig. 1 (symbols) for x = 0.05, together with the rescaled susceptibility (solid curve). By contrast, for x ≈ 0.1 the muon relaxation (not shown) becomes extremely fast already above the superconducting Tc. In polycrystals this is the signature of magnetic order taking place at TM > Tc [9]. A coexisting flux line lattice is hidden by the large magnetic damping, preventing its investigation by TF μSR. We characterized...
the superconducting response by dc magnetic susceptibility \( \chi(T) \) (inset of Fig. 1), measured by a superconducting quantum interference device (SQUID). Comparison with the bulk case, \( x = 0.05 \), allows us to roughly estimate the SC volume fraction of the other samples, which becomes absolutely marginal above \( x = 0.5 \), confirming previous reports [8]. The shielding response is indeed observable, but already much weaker for all samples with a reduced \( T_c \), at odds with the behavior of analogous Nd-1111 and La-1111 Ru-doped samples [10,11], suggesting that superconductivity is not a homogeneous bulk phenomenon in SmFe_{1−x}Ru_xAsO_{0.85}F_{0.15} for \( x \geq 0.1 \).

We now turn to zero-field (ZF)-\( \mu \)-SR results, sensitive to short-range magnetic order, owing to the short-range muon-spin coupling with the electronic moments. The time evolution of the (ZF)-\( \mu \)-SR asymmetry is displayed in the inset of Fig. 2 for \( x = 0.20 \) at three representative temperatures. Below a mean transition temperature \( T_M = 30 \) K the muon asymmetry is \( A_{ZF}(t) = a_{ZF}(w_T e^{-\sigma_T t^2/2} + w_L e^{-\lambda_L t}) \) displaying both a fast \( \sigma_T \) and a slow \( \lambda_L \) decay, where \( a_{ZF} \) is the high \( T \) value and \( w_T + w_L = 1 \). The two components reflect the orientation of the internal field with respect to the initial muon spin \( S_\mu \) (transverse, \( B_\perp \parallel S_\mu \), weight \( w_T \leq 2/3 \), and longitudinal, weight \( w_L \), \( B_\parallel \parallel S_\mu \) plus, possibly, a fraction with vanishing internal fields). The very fast transverse relaxation (\( \sigma_T \sim 30 \) ms\(^{-1} \)) represents the signature of a sizeable distribution of internal fields \( B_\parallel \) with standard deviation \( \Delta B_\mu = \sigma_T / \gamma = (B_\parallel^2)^{1/2} \) reaching 45 mT at low \( T \) (Fig. 2). This is a typical Fe dipolar field value at the muon site in F-doped 1111 close to a \( M \)-SC crossover [3,4]. The transverse component is overdamped down to 1.5 K (no asymmetry oscillations in Fig. 2, inset) and its fast relaxation is partially quenched in fields of order \( B_\perp = 50 \) mT applied along the initial muon-spin direction (not shown), as expected for a static \( B_\perp \). Therefore the overdamped muon-spin precessions are due to inhomogeneous magnetic short-range order [4].

The fraction of the sample volume where muons experience a net field \( B_i \) is calculated as \( V_M = 3w_T/2 = 3(1 - w_L)/2 \) [4]. Its dependence on temperature is shown in Fig. 2 for \( x = 0.20 \). It is noteworthy that \( V_M \) is 100% already below 20 K; i.e., internal fields develop throughout the whole sample well above the ordering temperature of the Sm sublattice \( (T_N^S \approx 5 \) K) [3,12] confirming that the observed magnetic order comes from the FeAs layers. In summary, since in all samples with \( x \geq 0.10 \) every muon detects a local magnetic field, every muon site must still be extremely close to some ordered magnetic moment. On the other hand in the same samples SQUID susceptibility detects a non-negligible shielding fraction. A similar scenario is found in cuprates and pnictides [3–5,13,14] and it may originate from magnetic and superconducting interspersed regions of nanometric size, compatible with proximity and the very short coherence length \( \xi \) of nonconventional superconductors [4]. Muons will measure a net field everywhere inside the nanoscopic mixture of \( M \) and SC regions, if these three length scales are comparable: the coherence length \( \xi \), the mean SC domain size \( d \) and the decay distance \( r \) of the dipolar field \( B_i \) from the closest \( M \) cluster (typically a few nm).

The complete phase diagram for SmFe_{1−x}Ru_xAsO_{0.85}F_{0.15}, obtained by combining the SQUID and the ZF-\( \mu \)-SR data, is shown in Fig. 3. At small Ru content a reentrant magnetic order within the FeAs layers is observed at the same \( x \sim 0.10 \), where \( T_c \) is dramatically reduced. Magnetic order in pnictides requires an orthorhombic structure, which has been recently demonstrated to occur [15] below 150 K already in our starting material, SmFeAsO_{0.85}F_{0.15}. Figure 3 shows also that, at high doping levels, both \( M \) and SC states disappear around the same threshold value \( x_c \sim 0.6 \), which is characteristic of magnetic interactions with nearest and next-nearest neighbors on a square lattice [16]. Notice that the same critical Ru content \( x_c \sim 0.6 \) is required to disrupt both superconductivity in REFe_{1−x}Ru_xAsO_{0.85}F_{0.11} with RE = La,
Nd [10,11] and magnetic order in fluorine free REFe$_{1-x}$Ru$_x$AsO with RE = La, Pr [17,18]. On one hand this common critical Ru threshold emphasizes the intimate relation between magnetism and superconductivity. On the other hand the degradation of SC when $M$ is recovered indicates a strong competition between the two respective order parameters. This apparent incongruity can be resolved by assuming that, in agreement with the most popular view, superconductivity is assisted by magnetic fluctuations. These fluctuations are suppressed by the full magnetic order, which competes with the SC state. Still short-range $M$ order may allow fluctuations to survive alongside the $M$ clusters, in nanoscopic form, and SC may survive as well. Upon approaching $x_c = 0.6$ both the static magnetism and the relevant magnetic fluctuations vanish, together with the superconductivity. Hence our results provide strong evidence for a superconductivity pairing of magnetic origin.

The presence of magnetic order in optimally F-doped RE1111 compounds was never observed before, and it is very surprising that it is induced by Ru substitution, iso-electronic to Fe and diamagnetic [8,17,18]. This scenario bears some analogies with that observed in cuprates, where the iso-electronic and diamagnetic Cu:Zn substitution recovers a frozen spin configuration, producing a concomitant sharp suppression of superconductivity [19,20].

Lang et al. [5] have shown that in F-doped 1111 two As local environments appear, as witnessed by two NQR peaks. They account for an intrinsic bimodal electronic inhomogeneity on a nanoscopic scale within the FeAs layers, being associated [5] to a charge-poor and a charge-rich local environment, respectively. NQR maps the local electronic configuration via the electric-field gradient at the $^{75}$As nucleus and we report here how its bimodal electric-field gradient is affected by Ru substitution. In Fig. 4(a) we show the NQR spectra for representative SmFe$_{1-x}$Ru$_x$AsO$_{0.85}$F$_{0.15}$ samples, together with the $x = 0$ data from Ref. [5]. In addition to the double peak structure (which is still visible, although much less pronounced than in F-underdoped samples [5]), up to four new satellites appear, each characterized by the number of Ru NN ions in the local fourfold coordination around As. Each spectrum has been fitted to a number of equal width Gaussian curves, varying from two to six. The five nearly equally spaced peaks of the $x = 0.56$ sample ($j = 0–4$) may be initially assigned to configurations with $j$ Ru NN ions, respectively.

Figure 4(b) displays the best-fit central frequency of the different peaks as a function of Ru content. They can be easily grouped into six families indicated by the straight lines. Figure 4(c) displays the weight of these peaks as a function of Ru content, measured as the relative area $a_j = A_j/A_{\text{Tot}}$ ($A_{\text{Tot}} = \sum_j A_j$ is the total area), proportional to the number of $^{75}$As nuclei in that environment. The $j = 0$ weight is taken as the sum of the two lower-frequency peaks, labeled 0A and 0B. Indeed Fig. 4(d) shows their separate concentration dependence, with a remarkable initial correlation: one collapses while the other grows with increasing Ru content. This suggests that they are two components of the same $j = 0$ Ru-free configuration. The assignment is supported by the good agreement of $a_j(x)$ with the fit to a binomial distribution, corrected with a weight redistribution that favors larger $j$, indicating a tendency towards Ru clustering [Fig. 4(c) solid lines].
dependence of both charge-poor environment (weight redistribution due to Ru. The presence of Ru favors the transition must be influenced directly by a

\[ j \]

Ru substitution in optimally F-doped \( \text{SmFe}_{1-x}\text{Ru} \), \( \text{AsO}_{0.35}\text{F}_{0.15} \) leads to a reentrant static magnetic order which degrades the superconducting ground state. The two order parameters compete, producing a nanoscopic phase separation. The two weights, tuned by the average Ru doping, scale with the corresponding transition temperatures. Both magnetism and superconductivity are suppressed at the percolation threshold characteristic of the magnetic system, suggesting that superconductivity cannot exist if magnetism is definitely suppressed by magnetic dilution. This picture strongly supports magnetic coupling models of superconductivity.

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The average effect of Ru substitution on the electronic properties is directly reflected in the weight of the two low frequency peaks, \( a_{0A}, a_{0B} \), around 14.5 MHz and 13 MHz respectively, [Fig. 4(b)], representing nuclei without NN Ru ions. Comparison with the \( x = 0 \) spectrum in Fig. 4 clarifies that this is the same doublet already present at \( x = 0 \) and discussed in Ref. [5] in terms of the electronic inhomogeneity of underdoped SC. Their symmetric anti-correlated behavior for \( x < 0.1 \), and the clear kinks for \( x = 0.1 \) are clearly related to the change in the magnetic and superconducting properties of the system.

This is confirmed in Fig. 5, showing a nearly linear dependence of both \( T_c \) and \( T_M \) when plotted vs the NQR weights \( a_{0A} \) and \( a_{0B} \), respectively. This correlation provides compelling evidence that the volume fraction of the charge-rich (charge-poor) local environments in the FeAs layers increases together with the strength of the average SC coupling \( (M \text{ coupling}) \), i.e., the transition temperature \( T_c (T_M) \). The relation between the average coupling and the spatial extension of the ordered phase suggests a percolation transition for both orders. Given that the total charge doping is nearly constant as a function of isoelectronic Fe:Ru substitution in 1111 compounds [8,17,18], the transition transition for both orders. Given that the total charge

\[ T_c \]

**FIG. 5** (color online). \( T_c \) and \( T_M \) versus the NQR weights \( a_{0A} \) and \( a_{0B} \) respectively with their linear regressions.

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