Superconductivity and Field-Induced Magnetism in SrFe$_{1.75}$Co$_{0.25}$As$_2$

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Using muon-spin rotation, we studied the in-plane ($\lambda_{ab}$) and the out of plane ($\lambda_c$) magnetic field penetration depth in SrFe$_{1.75}$Co$_{0.25}$As$_2$ ($T_c \approx 13.3$ K). Both $\lambda_{ab}(T)$ and $\lambda_c(T)$ are consistent with the presence of two superconducting gaps with the gap to $T_c$ ratios $2\Delta/k_BT_c = 7.2$ and $2.7$. The penetration depth anisotropy $\gamma_\lambda = \lambda_c/\lambda_{ab}$ increases from $\gamma_\lambda \approx 2.1$ at $T_c$ to 2.7 at 1.6 K. The mean internal field in the superconducting state increases with decreasing temperature, just opposite to the diamagnetic response seen in magnetization experiments. This unusual behavior suggests that the external field induces a magnetic order which is maintained throughout the whole sample volume.

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The discovery of Fe-based high-temperature superconductors (HTS) with critical temperatures above 50 K has triggered a surge of theoretical and experimental studies. Similar to the cuprates, the Fe-based HTS are characterized by a layered structure and by superconducting states which appear upon doping from parent compounds exhibiting long-range antiferromagnetic (AF) order. On the other hand, the metallicity of the parent compounds, as well as the occurrence of superconductivity on a few disconnected pieces on the Fermi surface clearly distinguish them from cuprate HTS. A fundamental question is to understand whether the mechanism(s) leading to the occurrence of the superconducting ground states in both families share common ground. On this frame, one of the most topical issues is to compare the interplay between magnetism and superconductivity between both families. This can be realized by carefully monitoring the emergence of these ground states as a function of chemical doping, applied magnetic field or external pressure.

In lightly doped cuprate HTS, static magnetism is found to coexist with superconductivity on a nanometer scale, as revealed by neutron diffraction, nuclear quadrupolar resonance and muon-spin rotation ($\mu$SR) experiments. For this family, stripe-like modulations of the charge and spin densities are found. On the other hand, for the Fe-based HTS REO$_{1-x}$F$_x$FeAs (RE=La, Ce, etc.) and (Sr,Ba)$_{1-x}$K$_x$Fe$_2$As$_2$ the majority of the reported phase diagram studies show either the occurrence of an abrupt first-order like change, with a full suppression of the AF order, at the doping where superconductivity emerges, or point to a microscopic separation of magnetism and superconductivity. To date, a microscopic coexistence of both states is solely reported for the system SmFeAsO$_{1-x}$F$_x$. e.g., even a magnetic field as low as 10 mT is sufficient to enhance the weak AF ordering over the entire sample volume. For Fe-based HTS we are aware of only two reports pointing to a possible enhancement of magnetism by applied magnetic field. Here, we report on $\mu$SR studies of a single crystalline sample of SrFe$_{1.75}$Co$_{0.25}$As$_2$. In zero external field, superconductivity coexists with dilute Fe spins which are static on the $\mu$SR time scale. In the superconducting state, the applied field leads to appearance of an additional source of local magnetic field, maintained throughout the whole volume of the sample, thus pointing to the field-induced ordering of the Fe moments.

The SrFe$_{1.75}$Co$_{0.25}$As$_2$ (SFCA) single crystalline samples were synthesized as described in[15]. The zero-field cooled (ZFC) and field cooled (FC) magnetization ($M$) measurements confirm bulk superconductivity (see Fig. 1 and Ref. 15). The transition temperature ($T_c$) of 13.3(1) K is consistent with the overdoped composition of the sample[16]. The true FC Meissner effect is very small, suggesting that pinning in SFCA is relatively strong. The ZFC shielding is 97–100%. All crystals used in our study were taken from the same growth batch and show very similar $M(T)$ dependences.

The $\mu$SR experiments were carried out at the $\pi$M3 beam line (Paul Scherrer Institute, Villigen, Switzerland). Three SFCA single crystals with an approximate size of $2.5 \times 2.0 \times 0.1$ mm$^3$ were mounted on a holder designed to conduct $\mu$SR measurements on thin single-crystalline samples. The zero-field (ZF) and transverse-field (TF) $\mu$SR experiments were performed at temperatures ranging from 1.5 to 150 K. In two sets of TF measurements the external magnetic field $\mu_0H = 10$ mT was applied parallel and perpendicular to the crystallographic $c$ axis, and always perpendicular to the muon-spin polarization. The typical counting statistics were $\sim 1.5 \cdot 10^7$ positron events for each particular data point.

In ZF, the muon-spin polarization is relaxed by magnetic moments of electronic and nuclear origin. As shown
in Fig. 2, the relaxation rate of the ZF-μSR signal is constant down to $T \simeq 5$ K, while at lower temperatures an additional fast relaxing component starts to develop. The solid lines in Fig. 2 correspond to the fit by:

$$A^{ZF}(t) = A_1 \exp(-\Lambda_1 t) + A_2 \exp(-\Lambda_2 t)$$  \hspace{1cm} (1)

Here $A_1$ and $A_2$ are the initial asymmetries of the slow and the fast relaxing components, and $\Lambda_1$ and $\Lambda_2$ are the corresponding exponential depolarization rates. The temperature dependence of $A_1$ normalized to the total ZF asymmetry $A_1 + A_2$ is shown in the inset. The fit reveals that $\Lambda_1$ is temperature independent which is also clearly seen from the raw data as a parallel shift of $A^{ZF}(t)$ at $t \gtrsim 0.2 \mu$s with decreasing temperature. Measurements in a longitudinal-field geometry indicate that the exponential character of the muon-spin relaxation is due to randomly oriented local magnetic fields, which are static on the μSR time scale. Such behavior is consistent with dilute Fe moments as observed recently for another representative of Fe-based HTS FeSe$_{0.85}$ [17].

In the TF geometry muons measure the magnetic field distribution $|P(B)|$ inside the sample. For the superconductor in the vortex state, $P(B)$ is uniquely determined by the magnetic penetration depth $\lambda$ and the coherence length $\xi$ [18]. Few representative $P(B)$ distributions, obtained after Fourier transformation of TF μSR time-spectra, are presented in Figs. 2b, c. In the normal state, a symmetric line at the position of the external magnetic field with a broadening arising from the nuclear and electronic magnetic moments is seen. Below $T_c$, the field distribution is broadened and asymmetric which is characteristics of the inhomogeneous field distribution within the flux line lattice (FLL).

In an orthogonal reference frame $x, y, z$ with $H \parallel z$ ($z$ is one of the principal axes $a, b, \text{or } c$) the spatial magnetic field distribution within a FLL of an anisotropic superconductor is [18]:

$$B(r) = \langle B \rangle \sum_G \exp(-i \mathbf{G} \cdot \mathbf{r}) B_G(\lambda_x, \lambda_y, \xi_x, \xi_y).$$  \hspace{1cm} (2)

$\mathbf{G}$ is the reciprocal lattice vector, $\langle B \rangle$ is the average magnetic field inside the superconductor, $\mathbf{r}$ is the vector coordinate in a plane perpendicular to the applied field, and the Fourier components $B_G$, obtained within the framework of the Ginzburg-Landau (GL) model, are [18]:

$$B_G = \frac{\Phi_0}{S} (1 - b^4) \frac{uK_1(u)}{\lambda_2 G_{G_y}^2 + \lambda_2 G_{G_z}^2}.$$  \hspace{1cm} (3)

Here, $\Phi_0$ is the magnetic flux quantum, $S = \Phi_0/\langle B \rangle$ is the FLL unit cell area, $b = \langle B \rangle/B_{c2}$ ($B_{c2}$ is the second critical field), $K_1(u)$ is the modified Bessel function, and $u^2 = 2(\xi_2^2 G_{G_x}^2 + \xi_0^2 G_{G_z}^2)(1 + b^4)(1 - 2b(1-b)^2)$. The reciprocal lattice corresponding to the field distribution in hexagonal FLL is: $G_{m,n} = [(2\pi/S)[ym, (n - m/2)x]$, where $x = (2\lambda x/\sqrt{3}\lambda y)^{1/2}$, $y = (\sqrt{3}S\lambda y/2\lambda z)^{1/2}$, and $m$ and $n$ are the integer numbers [19]. Note that in the uniaxial case ($\lambda_x = \lambda_y$), Eq. (3) converts into the standard GL equation for an isotropic superconductor [18].
The TF μSR time spectra were fitted to a theoretical asymmetry function $A_T^T(t)$ by assuming the internal field distribution $P_{\text{id}}(B)$ obtained from Eq. 2 and accounting for the FLL disorder and the electronic moment contributions (see the ZF discussion above) by convoluting $P_{\text{id}}(B)$ with Gaussian and Lorenzian functions:

$$A_T^T(t) = A e^{i\phi} e^{-\sigma^2 r^2 t^2 / 2 - \lambda t} \int P_{\text{id}}(B) e^{i\gamma B t} dB.$$ (4)

Here $A$ and $\phi$ are the initial asymmetry and the phase of the muon spin ensemble, $\sigma_g$ accounts for the FLL disorder 21, and $\Lambda$ relates to the electronic moment contribution and is assumed to be equal to $A_1$ obtained in ZF experiments. During the analysis we first fit the data measured in $H \parallel c$ orientation which, considering $\lambda_0 = \lambda_b$, allows to obtain the in-plane magnetic penetration depth $\lambda_{ab}$. Note that at such a low field as $\mu_0 H = 10$ mT, $P(B)$ is independent on $\xi$ 20. $\lambda_b$ was further used in the fit of $H \perp c$ set of data by assuming in Eq. 3 $\lambda_x = \lambda_{ab}$ and $\lambda_y = \lambda_c$. The resulting fitted curves are represented in Figs. 2b and c by red lines. The field distributions obtained by Eq. 2 are shown in the insets.

The temperature dependences of the initial asymmetries are shown in the inset of Fig. 2a. Both, $A_{\parallel c}(T)$ and $A_{\perp c}(T)$ are almost constant down to $T \approx 3$ K and decrease by $\approx 10\%$ at $T = 1.6$ K just resembling the temperature behavior of the slow relaxing component $A_1$ observed in ZF measurements. This suggests that the fast relaxing component, seen in ZF μSR experiments, appears from areas which are separated in space from the “slow-relaxing” ones. The superconductivity in such magnetic regions may either coexist with magnetism on a nanometer scale 10, or become suppressed due to enhanced magnetic order 8, 21.

The dependence of $\lambda_{ab}^{-2}$ and $\lambda_c^{-2}$ on temperature is shown in Fig. 3a. The experimental data were analyzed within the framework of the phenomenological $\alpha$-model by assuming two independent contributions to $\lambda^{-2}$ 22:

$$\frac{\lambda^{-2}(T)}{\lambda^{-2}(0)} = \omega \cdot \frac{\lambda^{-2}(T, \Delta_{0,1})}{\lambda^{-2}(0, \Delta_{0,1})} + (1 - \omega) \cdot \frac{\lambda^{-2}(T, \Delta_{0,2})}{\lambda^{-2}(0, \Delta_{0,2})}$$ (5)

In this model, each superconducting gap, $\Delta_1(T)$ and $\Delta_2(T)$, has a similar temperature dependence given by $\Delta_i(T) = \Delta_{i,0} \tanh[(1.82[1.018(T_c/T - 1)]^{0.51}$. Ref. 22, but different zero-temperature values ($\Delta_{0,i}, i = 1, 2$). In our analysis both the large and the small gap were assumed to be of $s$-wave symmetry. The parameter $\omega$ accounts for the relative contribution of the larger gap to $\lambda^{-2}$ and $\lambda^{-2}(0)$ is the penetration depth at $T = 0$. Each component $\lambda^{-2}(T, \Delta_{0,i})/\lambda^{-2}(0, \Delta_{0,i})$ is calculated within the local (London) approximation 23. During the fit, both $\lambda_{ab}(T)$ and $\lambda_c(T)$ were assumed to be described by the same small and large gaps ($\Delta_{1,ab} = \Delta_{1,c}$ and $\Delta_{2,ab} = \Delta_{2,c}$), but different weighting factors ($\omega_{ab} \neq \omega_c$). The results of the fit are summarized in Fig. 3a and Ta-

**FIG. 3:** (Color online) (a) Dependence of $\lambda_{ab}^{-2}$ and $\lambda_c^{-2}$ on temperature of SrFe$_{17}$Co$_{0.25}$As$_2$. The solid lines are the fits by Eq. 5 with the parameters summarized in Table II. The inset shows the temperature dependence of the penetration depth anisotropy $\gamma_\lambda = \lambda_c/\lambda_{ab}$. (b) Temperature dependence of $\langle B \rangle - \langle B \rangle^{\text{ns}}$ obtained in $H \parallel c$ (red symbols) and $H \perp c$ (blue symbols) set of experiments. The inset shows the dependence of $\langle B \rangle(1.6 \text{K}) - \langle B \rangle^{\text{ns}}$ on $\langle B \rangle^{\text{ns}}$ in $H \parallel c$ orientation.

**Table II** The temperature dependence of the anisotropy parameter $\gamma_\lambda = \lambda_c/\lambda_{ab}$ is shown in the inset. Four important results are deduced from this analysis: (i) The contribution of the large gap to $\lambda_{ab}^{-2}(T)$ is relatively small in comparison with that observed in Ba$_{1-x}$K$_x$Fe$_2$As$_2$ 21. This may be explained by the fact that the electron doping due to Co substitution reduces the size of the Fermi surface pockets at the Brillouin zone center (where the large gap opens), but leads to substantial enhancement of the pockets at the zone corner (where the small gap develops) 24, 25. (ii) $\gamma_\lambda$ increases from $\gamma_\lambda \approx 2.1$ at $T_c$ to 2.7 at 1.6 K. This increase is consistent with the general trend obtained for various Fe-based HTS 21, 26. Our data imply that the different temperature dependences of $\lambda_{ab}^{-2}$ and $\lambda_c^{-2}$, as well as dependence of $\gamma_\lambda$ on $T$, are due to much smaller contribution of the larger gap to $\lambda_{ab}^{-2}$ than that to $\lambda_c^{-2}$. (iii) $\gamma_\lambda$ is very close to the calculated ratio of the plasma frequencies $\gamma_\omega = \omega_{p,ab}/\omega_{p,c} \approx 2.8$ of Sr$_2$Fe$_2$As$_2$. 27. Note that within the London theory $\gamma_\lambda \equiv \gamma_\omega$. (iv) The values $2\Delta_{0,1}/k_B T_c = 7.2$ and $2\Delta_{0,2}/k_B T_c = 2.7$ are well within the ranges established for various Fe-based HTS. As shown in Ref. 27, Fe-based HTS bear two nearly isotropic gaps with $2\Delta/k_B T_c = T \pm 2$ and $2.5 \pm 1.5$. 


TABLE I: Summary of \(\lambda \) magnetic field dependence of \(\{B\}^{\text{ns}}\) study of SrFe\(_{1.75}\)Co\(_{0.25}\)As\(_2\). (see text for details).

| \(T_c\) (K) | \(\Delta_{0,1}\) (meV) | \(\Delta_{0,2}\) (meV) | \(\omega\) (as) | \(\lambda(0)\) (nm) |
|-----------|-----------|-----------|-------------|-----------------|
| 13.35 \(^\circ\) | 4.14 \(^\circ\) | 7.2 | 1.56 \(^\circ\) | 2.7 | 0.04 | 315 |
| 13.35 \(^\circ\) | 4.14 \(^\circ\) | 7.2 | 1.56 \(^\circ\) | 2.7 | 0.29 | 870 |

*Common parameters used in the analysis.

In Figure 3b we plot the difference between the internal magnetic field \(\langle B \rangle\) and that measured in the normal state at \(T \approx 20\) K \((\{B\}^{\text{ns}})\). In contrast to what is expected for a superconductor, \(\langle B \rangle\) increases with decreasing temperature. We may clearly rule out the possibility to explain the positive field shift by a paramagnetic Meissner effect, since both ZFC and FC magnetization result in a diamagnetic shift (see Fig. 1). Also, it cannot be explained since both ZFC and FC magnetization result in a diamagnetic shift (see Fig. 1). Also, it cannot be explained by the reduction of a hypothetical negative muon Knight shift due to condensation of the carriers into Cooper pairs 28, otherwise the difference between \(\langle B \rangle\) \((1.6\) K\), measured deeply in the superconducting state, and \(\{B\}^{\text{ns}}\) would increase with increasing field. The inset in Fig. 3b shows, in contrast, that \(\langle B \rangle\) \((1.6\) K\) \(- \{B\}^{\text{ns}}\) decreases.

Both, the temperature and the magnetic field dependence of \(\langle B \rangle - \{B\}^{\text{ns}}\) resemble the situation in the electron doped cuprate HTS \(Pr_{2-x}Ce_xCuO_4\) 11 and \(Pr_{1-x}LaCe_xCuO_4\) 12. Those reports are different in some details, but agreed that the paramagnetic response in the superconducting state, seen by muons, is caused by the field-induced weak AF order. The external field applied in \(c\) direction leads to the appearance of an additional field component at the muon stopping site, which is parallel to the \(c\) axis 11, 12. We may assume that the paramagnetic response of SFCA, seen in Fig. 3b, is caused by the same mechanism. The solid line in the inset of Fig. 3b corresponds to the fit by \(\langle B \rangle \sim \frac{2\Delta_{0,1}^2 + 2\Delta_{0,2}^2}{\Delta_{0,1}^2 + \Delta_{0,2}^2} \langle B \rangle\) \((1.6\) K\) \(- \{B\}^{\text{ns}}\) with the induced field component \(B^\perp\) \(= 5.2\) mT. We emphasize, that the field-induced AF order in SFCA is different from that for the parent compound SrFe\(_2\)As\(_2\), where the magnetic field on the muon site is parallel to the \(c\) axis 24.

The strong influence of the superconducting phase is observed at external field as low as \(10\) mT, which corresponds to the intervortex distance \(\sim 500\) nm. Considering that most of the implanted muons probe the sample outside of the vortex cores, we may conclude that the field-induced AF order extends throughout the whole sample volume. Further experimental and theoretical studies are needed in order to elucidate the origin of these effects, as well as to consider a possible role of muons as a source of perturbation.

To conclude, muon-spin rotation measurements were performed on a single-crystalline sample of SrFe\(_{1.75}\)Co\(_{0.25}\)As\(_2\) \((T_c \approx 13.3\) K\). In zero field, superconductivity coexists with dilute Fe moments which are static on the \(\mu\)SR time scale. The temperature dependence of the in-plane \(\lambda_b\) and the out of plane \(\lambda_c\) magnetic penetration depth are well described assuming the presence of two \(s\)-wave like gaps with zero-temperature values \(\Delta_{0,1} = 4.14\) meV and \(\Delta_{0,2} = 1.56\) meV, respectively. The gap to \(T_c\) ratios are \(\frac{2\Delta}{k_BT_c} = 7.2\) and 2.7, in agreement with those reported for various Fe-based HTS 27. The mean internal field in the superconducting state increases with decreasing temperature, just opposite to the diamagnetic response seen in magnetization experiments. This may be due to weak field-induced antiferromagnetic order leading to appearance of in-plane magnetic field component on the muon stopping site. The fact that the magnetism is induced by a field as low as \(10\) mT points to strong interplay between the magnetic and the superconducting order parameters in Fe-based HTS. Whether these order parameters coexist or separate on a microscopic level is still an open question.

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1. S. Watanabe et al., Phys. Rev. B 68, 172501 (2003).
2. M.H. Julien et al., Phys. Rev. Lett. 83, 604 (1999).
3. Ch. Niedermayer et al., Phys. Rev. Lett. 80, 3843 (1998).
4. S. Sanna et al., Phys. Rev. Lett. 93, 207001 (2004).
5. H. Luetkens et al., Nature Materials (2009), DOI: 10.1038/nmat2397.
6. J. Zhao et al., Nature Materials 7, 953 (2008).
7. M. Rottet et al., Angew. Chem. Int. Ed. 47, 7949 (2008).
8. T. Goko et al., arXiv:0805.1425 (unpublished).
9. A.J. Drew et al., Nature Materials (2009), DOI: 10.1038/nmat2396.
10. S. Sanna et al., arXiv:0902.2156 (unpublished).
11. J.E. Sonier et al., Phys. Rev. Lett. 91, 147002 (2003).
12. R. Kadono et al., J. Phys. Soc. Jpn. 73, 2944 (2004); R. Kadono et al., J. Phys. Soc. Jpn. 74, 2806 (2005).
13. H. Luetkens et al., Phys. Rev. Lett. 101, 097009 (2008).
14. P.L. Pratt et al., Phys. Rev. B 79, 052508 (2009).
15. D. Kasinathan et al., New J. Phys. 11, 052023 (2009).
16. A. Leithe-Jasper et al., Phys. Rev. Lett. 101, 207004 (2008).
17. R. Khasanov et al., Phys. Rev. B 78, 220510 (2008).
18. A. Yaouanc et al., Phys. Rev. B 55, 11107 (1997).
19. S.L. Thiemann et al., Phys. Rev. B 39, 11406 (1989).
20. A. Maisuradze et al., J. Phys.: Condens. Matter 21, 075701 (2009).
21. R. Khasanov et al., arXiv:0901.2329 (unpublished).
22. A. Carrington and F. Manzano, Physica C 385, 205 (2003).
23. R. Khasanov et al., Phys. Rev. B 77, 184512 (2008).
24. Y. Sekiba et al., arXiv:0812.4111 (unpublished).
25. K. Terashima et al., arXiv:0812.3704 (unpublished).
26. S. Weyeneth et al., arXiv:0811.4047 (unpublished); M.A. Tanatar et al., arXiv:0812.4991 (unpublished).
27. D.V. Evtsushinsky et al., submitted to New J. Phys.
28. R. Feyerherm et al., Phys. Rev. Lett. 73, 1849 (1994).
29. H. Luetkens et al. under preparation.