Coexistence of magnetic fluctuations and superconductivity in the pnictide high temperature superconductor \( \text{SmFeAsO}_{1-x}\text{F}_x \) measured by muon spin rotation

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Muon-spin rotation experiments were performed on the pnictide high temperature superconductor \( \text{SmFeAsO}_{1-x}\text{F}_x \) with \( x = 0.18 \) and 0.3. We observed an unusual enhancement of slow spin fluctuations in the vicinity of the superconducting which suggests that the spin fluctuations contribute to the formation of an unconventional superconducting state. An estimate of the in-plane penetration depth \( \lambda_{ab}(0) = 190(5) \text{nm} \) was obtained, which confirms that the pnictide superconductors obey an Uemura-style relationship between \( T_c \) and \( \lambda_{ab}(0)^{-2} \).

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The recent discovery of high temperature superconductivity (HTSC) in the layered tetragonal pnictide compound \( R\text{FeAsO}_{1-x}\text{F}_x \) (\( R=\text{La}, \text{Nd}, \text{Pr}, \text{Gd}, \) and \( \text{Sm} \)) has been tracked as a function of F-doping. These measurements suggest that the magnetic order disappears \[8\]. Recent neutron measurements on F-doped superconducting samples confirm this conjecture since they could not detect any magnetic order \[9\]. Thus an important issue is whether weak, slowly fluctuating or strongly disordered magnetism persists in these superconductors.

In this letter we report a \( \mu \)SR study which provides new insight into the magnetic properties of this new superconductor. Two polycrystalline samples with nominal compositions of \( x = 0.18 \) and 0.3 were synthesized by conventional solid state reaction methods as described in Ref \[2, 8\]. Standard powder x-ray diffraction patterns were measured where all (the main) peaks could be indexed to the tetragonal \( \text{ZrCuSiAs} \)-type structure for \( x = 0.18 \) (\( x = 0.3 \)), as previously reported \[2, 8\]. DC resistivity and magnetisation measurements were made to determine \( T_c \) (\( \Delta T_c = 45(3) \) and 45(4) K for \( x = 0.18 \) and 0.3 corresponding to the midpoint (10% to 90% width) of the resistive and the diamagnetic transitions.

The \( \mu \)SR experiments were performed at the EMU, MuSR and ARGUS instruments of the ISIS facility, Rutherford Appleton Laboratory, UK, which provides pulsed beams of 100% spin polarized muons. \( \mu \)SR measures the time evolution of the spin polarization of the implanted muon ensemble using the time-resolved asymmetry \( A(t) \) of muon decay positrons. The technique \[11, 12\] is well suited to studies of magnetic and superconducting (SC) materials as it allows a microscopic determination of the internal field distribution and gives direct access to the volume fractions of SC and magnetic phases \[12\].

Figure 1(a) shows representative spectra of the zero-field (ZF) \( \mu \)SR measurements at three different temperatures for \( x = 0.18 \). The relatively fast relaxation of \( A(t) \), which persists even at 200 K, provides clear evidence for
we find that these spectra are well described with a single stretched exponential relaxation function of the form $A(t) = A(0)G(t)$ where the spin polarization function is $G(t) = \exp[-(\lambda_{ZF} t)^\beta]$. This is illustrated in the inset of Fig. 1(a) which shows that the data follow straight lines on a log-log plot of $-t/\ln(G(t))$ versus $t$ [13]. The temperature dependences of the relaxation rate, $\lambda_{ZF}$, and the exponent, $\beta$, are shown in Figs. 1(b) and 1(c), respectively. Above 100 K the relaxation is exponential with $\beta \approx 1$ and $\lambda_{ZF}$ is only weakly temperature dependent. Below 100 K the value of $\lambda_{ZF}$ exhibits a significant increase followed by a saturation below 30 K with a low temperature value of $\lambda_{ZF} \approx 1.2 \mu s^{-1}$. At the same time $\beta$ decreases continuously towards $\beta \approx 0.5$. Notably, the biggest changes occur in the vicinity of the SC transition at $T_C = 45(3)$ K, as shown by the vertical dashed line in Figs 1(b) and 1(c). The inset of Fig. 1(b) shows corresponding data for the $x=0.3$ sample where $\lambda_{ZF}$ is reduced but exhibits a similar temperature dependence.

In order to distinguish between static and dynamic contributions, a longitudinal field (LF) scan was performed at 60 K (Fig. 2). We observe an abrupt transition in the longitudinal relaxation rate $\lambda_{LF}$ at around 40 G but subsequent increases in $\lambda_{LF}$ produce no further significant change. This unusual behavior cannot be accounted for by a simple decoupling model for purely static or dynamic spins (dashed line in Fig. 2) which would predict $\lambda_{LF} \propto B^{-2}$ above some critical field (corresponding either to the internal field in the static case or $\nu/\gamma_\mu$ in the dynamic case, where $\nu$ is the fluctuation rate).

Two potential magnetic sources for the muon relaxation are: (1) the lanthanide moments in the SmO layers; (2) magnetic fluctuations originating from spin correlations in the FeAs layers. The strong doping dependence of the value of $\lambda_{ZF}$ (Fig. 1(b)) demonstrates clearly that the observed magnetism cannot be explained solely in terms of weakly coupled Sm moments. The data for $T > T_C$ naturally separates into two components: $T$-independent and $T$-dependent, as indicated by the guides to the eye in Fig. 1(b). The $T$-dependent component has an activation energy of 13(2) meV, a typical scale for lanthanide moment fluctuations [14]. The amplitude corresponds to the non-quenched component seen in LF and we ascribe this component to Sm moments which are fluctuating rapidly in this temperature region due to crystal field excitations [14]. The temperature-independent component which can be quenched in 40 G at 60 K can be identified with low energy spin fluctuation processes associated with the FeAs layer.

Although the high temperature relaxation is simple exponential and can therefore be associated with a single, dominant fluctuation rate, cooling through $T_C$ results in a reduction in $\beta$, signifying a range of fluctuation rates and/or local field amplitudes. This indicates that the spin dynamics at low temperature becomes substan-
tially more complex. Below $T_c$, the activated behavior ceases and $\lambda^{ZF}$ saturates, demonstrating that an additional relaxation channel becomes dominant. Although relaxation with $\beta = 0.5$ can be suggestive of glassy behavior or static disorder, it is unlikely to be the case here. Since it is known that the Sm moments order antiferromagnetically below $\sim 3 - 4\,K$ \cite{15} (and see below), we may preclude the possibility of the Sm moments forming a static disordered state at a higher temperature. While it is possible that static Fe moments could develop in this temperature regime, the lack of change in $\lambda^{ZF}$ in the region below 30 K where $\beta$ is changing continuously makes this interpretation unlikely. We can also discard any interpretation of our data which involves a small fraction of ordered spins, since the observed relaxation corresponds to the behavior of the overwhelming bulk of the sample across the entire temperature range; thus the role of any purported minority phase is not directly probed in these experiments.

Accordingly, our data suggest that the onset of superconductivity in this Sm compound is accompanied or slightly preceded by the enhancement of slow spin fluctuations which originate (at least partially) in the FeAs layers. This raises the question whether this coincidence is accidental or rather signifies that the spin fluctuations are playing an active role in the SC pairing mechanism. The former scenario is not supported by the similarities to (U,Th)Be$_2$13 and Sr$_2$RuO$_4$ \cite{21} where the beaking of time-reversal symmetry yields spontaneous supercurrents that create internal magnetic fields below $T_c$. However, these magnetic fields should be static rather than dynamic. Also the Sm moment would have to play an important role in this unconventional SC state since no corresponding increase in the ZF relaxation rate has been observed in the related La compound \cite{16,17}.

Certainly, our observations call for further investigations to clarify the role of slow spin fluctuations in the SC pairing mechanism and to explore whether the enhanced spin fluctuations in the Sm compound as compared to the La one are brought about by the coupling to the lanthanide moments or rather by the related structural changes \cite{1,2}.

Unconventional superconductivity is also supported by our transverse field TF-$\mu$SR measurements which yield a low temperature value of the in-plane magnetic penetration depth, $\lambda_{ab}(0)$, that falls close to the so-called Uemura-line of the cuprate HTSC \cite{22}. The TF-$\mu$SR spectra for $B_{app} = 100\,G$ were well described with a sum of two Gaussian functions using the form

$$A(t) = \sum_{i=1,2} A_i(0) \cos(\gamma_i B_t t) \exp \left[-\left(\frac{\sigma_{TF}^i}{2}\right)^2\right], \quad (1)$$

where $A_i$, $B_i$, and $\sigma_{TF}^i$ correspond to the amplitude, the local magnetic field at the muon site, and the relaxation rate, respectively. The second weakly-damped component reflects the small background from muons not stopping in the sample. The first, dominant component is due to the sample, and its temperature dependence is shown in Fig. 3. A sharp rise of $\sigma_{TF}$ [Fig. 3(a)] is seen below $T_c$ which exceeds that which would be expected from the ZF data; this additional contribution reflects the formation of the vortex lattice. This interpretation is confirmed by an observed diamagnetic shift of $\sim 13\,G$ [plotted in Fig. 3(b)] which also occurs at $T_c$. The additional steep rise of $\sigma_{TF}$ below 4 K [see inset to Fig. 3(a)] likely represents additional local-field broadening due to
the ordering of the Sm moments.

Notably, on cooling towards $T_c$, there is a gradual onset of a diamagnetic shift already at about 70 K, which is detailed in the inset to Fig.3(b). As was already noted above, this result could be an indication for a precursory SC state with a transition temperature higher than the bulk $T_c$ or the onset of SC fluctuations above $T_c$. However, at present we cannot rule out the possibility that the slowing down of the spin fluctuations leads to this reduction of the local field. In any case, the sharp onset of the diamagnetic shift at $T_c$ and the corresponding increase in $\sigma^{TF}$ allows us to provide an estimate of the in-plane magnetic penetration depth. Allowing for an additional root-exponential damping in the dominant term of (1) to take account of the contribution to the relaxation from magnetic fluctuations (which is known from the ZF measurements), the SC vortex contribution to the total linewidth $B_{\text{rms}}$ is obtained. This is plotted in Fig. 4(a) against applied field at 10 K and from this data we derive $\lambda_{ab} = 190(5)$ nm. This estimate is derived from fitting to the numerical results of a recent detailed Ginzburg-Landau vortex lattice calculation [22], taking a polycrystalline average for our powder sample in the high anisotropy limit, under the assumption that the length scales $\lambda$ and $\xi$ diverge following $1/\cos \theta$ as the field orientation approaches the plane at $\theta = 0$.

Since the estimate is made at $0.2 T_c$, it should provide a good account of $\lambda_{ab}(0)$, assuming a two-fluid type of saturating temperature dependence. Note that we are unable to establish whether any additional gap-node related linear term might be present at low temperatures due to the extra relaxation contribution below 4 K. Our value of $\lambda_{ab}$ is shorter than those found for LaFeAsO$_{1-x}$F$_x$ by Luetkens et al. (254(2) nm for $x = 0.1$ and 364(8) nm for $x = 0.07$ [16, 23]), reflecting the higher $T_c$ of our compound and hence larger superfluid stiffness (proportional to $\lambda_{ab}^2$). Fig. 4(b) shows the Uemura plot [24] for the high temperature pnictide superconductors measured to date by $\mu$SR. It appears that the overall trend lies closer to that of the hole-doped than the electron-doped cuprates.

In conclusion, the $\mu$SR results on polycrystalline SmFeAsO$_{1-x}$F$_x$ with $x=0.18$ and 0.3 provide clear evidence for the coexistence and interplay of superconductivity and dynamic magnetic correlations. The magnetic correlations exhibit a complex temperature dependence and a significant contribution of magnetic fluctuations to the enhanced $T_c$ is suggested. From TF measurements we obtained an estimate of the in-plane magnetic penetration depth of $\lambda_{ab} = 190(5)$ nm, which comes rather close to the Uemura line of the hole doped cuprates.

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Note added: Since submission of this Letter we learned of Ref. [26] which interprets the ZF-$\mu$SR of SmFeAsO$_{0.85}$ in terms of two exponential components and attributes the relaxation solely to weakly coupled Sm moments. Such an interpretation is inconsistent with our relaxation data and its LF and x dependences.

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\( x = 0.1 \) sample of Ref. 16 yields \( \lambda_{ab} = 254(2) \) nm and \( \kappa = 89(10) \) (Fig.4a), fully consistent with the \( \lambda_{ab} \) estimate of these authors, however our analysis shows that the field dependence of the linewidth is well described by a standard Ginzburg-Landau model (Ref.16).

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