# Field and Temperature Dependence of the Superfluid Density in $LaFeAsO_{1-x}F_x$ Superconductors: A Muon Spin Relaxation Study

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We present zero field and transverse field muon spin relaxation experiments on the recently discovered Fe-based superconductor $LaFeAsO_{1-x}F_x$ ($x = 0.075$ and $x = 0.1$). The temperature dependence of the deduced superfluid density is consistent with a BCS $s$-wave or a dirty $d$-wave gap function, while the field dependence strongly evidences unconventional superconductivity. We obtain the in-plane penetration depth of $\lambda_{ab}(0) = 254(2)$ nm for $x = 0.1$ and $\lambda_{ab}(0) = 364(8)$ nm for $x = 0.075$. Further evidence for unconventional superconductivity is provided by the ratio of $T_c$ versus the superfluid density, which is close to the Uemura line of high-$T_c$ cuprates.

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The ongoing search for new superconductors has recently yielded a new family of Fe-based compounds composed of alternating $La_2O_{2-x}F_x$ and $Fe_2As_2$ layers with transition temperatures $T_c$ up to 28 K [1]. By replacing La with other rare earths, $T_c$ can be raised to above 50 K [2], and thus the first non-copper-oxide superconductor with $T_c$ exceeding 50 K has emerged. Both recent experimental findings and theoretical treatments [3,4] indicate unconventional multiband superconductivity in the layers of paramagnetic Fe ions, which would normally destroy superconductivity in traditional $s$-wave superconductors. Point contact tunneling spectroscopy [5], specific heat [6], and magnetization measurements [7] point to nodal order parameters. High magnetic field experiments yielded evidence for two-band effects [8]. Various scenarios for superconductivity have also been discussed theoretically and different pairing symmetries of the superconducting ground state including spin-triplet $p$-wave pairing have been proposed [9–15]. Intriguingly, there is evidence for a close interplay between superconductivity and magnetism as it is well established for other unconventional superconductors. A commensurate spin-density wave (SDW) has been observed below 150 K in the undoped compound [16–19], and a recent theoretical work suggests that fluctuations associated with a magnetic quantum critical point are essential for superconductivity in the $F$-doped system [20].

In this Letter, we report zero field (ZF) and high transverse field (TF) muon spin relaxation measurements ($\mu$SR) on polycrystalline samples of $LaFeAsO_{1-x}F_x$ with $x = 0.075$ and 0.10. Our ZF-$\mu$SR experiments show that no static magnetic correlations are present down to 1.6 K. Hence, the spin-density state is completely suppressed upon $F$ doping. Properties of the superconducting state are determined by the TF-$\mu$SR measurements: The weak temperature dependence of the superfluid density below $T_c/3$ is consistent with both an $s$-wave and a dirty $d$-wave scenario. However, the field dependence of the static linewidth contradicts a standard BCS $s$-type gap function. In addition, our data provide the first quantitative measurements of the in-plane penetration depths, which amount to several hundred nanometers. This implies a dilute superfluid in $LaFeAsO_{1-x}F_x$ and, remarkably, the data for the Fe-based superconductors are very close to the Uemura line of the hole doped high-$T_c$ cuprates.

Polycrystalline samples of $LaFeAsO_{1-x}F_x$ ($x = 0.075, 0.1$) were prepared by using a two-step solid state reaction method, similar to that described by Zhu *et al.* [21], and annealed in vacuum. The samples consist of 1–100 $\mu$m sized grains of $LaFeAsO_{1-x}F_x$. The crystal structure and the composition were investigated by powder x-ray diffraction [$a = 4.026\,58(5)$, $c = 8.712\,32(15)$ for $x = 0.075$, and $a = 4.024\,51(6)$, $c = 8.709\,95(16)$ for $x = 0.1$] and wavelength-dispersive x-ray spectroscopy yielding $x_{WDX} = 0.065(10)$ and $x_{WDX} = 0.105(10)$, respectively.

In order to characterize the superconducting properties, zero field cooled (shielding signal) and field cooled (Meissner signal) magnetic susceptibility in external fields $H = 10$ Oe–50 kOe have been measured using a SQUID magnetometer. The resistance has been measured with a standard 4-point geometry employing an alternating dc current. Critical temperatures of $T_c \approx 26.0$ K and $T_c \approx 22$ K for $x = 0.1$ and $x = 0.075$, respectively, are extracted from these measurements (cf. Fig. 1).

In Fig. 2 we show representative ZF-$\mu$SR data for $x = 0.1$ at 1.6, 10, and 30 K. At all temperatures a weak Gaussian Kubo-Toyabe-like [22] decay of the muon spin polarization is observed. The relaxation rates are very small and can be traced back to the tiny magnetic fields

 

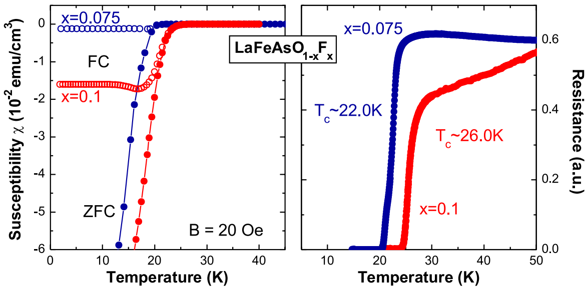


FIG. 1 (color online). Right: Temperature dependence of the resistance of $LaFeAsO_{1-x}F_x$ at $x = 0.1$ and $x = 0.075$ in the vicinity of $T_c$. Left: Field cooled and zero field cooled magnetic susceptibility for $x = 0.1$ and $x = 0.075$.

originating from nuclear moments. This implies that we can rule out any static SDW magnetism with considerable magnetic moments, for both the optimally and the underdoped samples. Thus, our data are in striking contradiction with the prediction in Ref. [3]. Instead, our measurements show that doping the compound LaFeAsO with electrons suppresses the SDW instability, and simultaneously promotes the superconductivity as the new ground state. This indicates that the superconducting phase is close to a quantum critical point related to the magnetic instability.

Such a proximity of magnetism and superconductivity is also suggested by several aspects of our $\mu$SR data. The first weak hint is a slight increase of the relaxation rate below $T_c$; see Fig. 2. Further indications arise from the high-TF measurements on $LaFeAsO_{0.925}F_{0.075}$, as will be discussed below.

The results of the analysis of the magnetic field dependent and temperature dependent TF-$\mu$SR measured in the vortex phase $H > H_{c1} \approx 40$ Oe are displayed in Figs. 3 and 4. From the muon spin polarization $P(t)$ we determined the Gaussian relaxation rate $\sigma$ which is the sum of a nuclear contribution $\sigma_{nm}$ and a contribution $\sigma_{sc}$ proportional to the second moment of the magnetic field distribution of the vortex lattice, i.e., $\sigma^2 = \sigma_{sc}^2 + \sigma_{nm}^2$. In order to extract the $\sigma_{sc}$ which measures the superfluid density, i.e., $\sigma_{sc}^2 \propto 1/\lambda^2 \propto n_s/m^*$ [23], we determined the small nuclear relaxation rate $\sigma_{nm} = 0.07(1)$ MHz at 30 K for both samples.

Figure 3 shows the obtained field dependence of $\sigma_{sc}$ for both $LaFeAsO_{1-x}F_x$ samples at 1.6 K. Each measurement was performed after cooling the sample in the field from above $T_c$.

We restrict our discussion of $\sigma_{sc}(H)$ to the optimally doped sample since the data for $x = 0.075$ are clearly influenced by additional contributions. In particular, after a small decrease we find that $\sigma_{sc}(H)$ starts to increase with increasing field, which is incompatible with the suppression of the superfluid density in high magnetic fields. Analogous behavior has been observed in high-$T_c$ cuprates [24,25], where an external field can promote the magnetic correlations leading to spurious magnetism in the otherwise superconducting sample. The upturn of $\sigma_{sc}$ as a function of magnetic field is therefore most likely produced by an additional magnetic contribution and does not reflect a field dependence of the superfluid density.

In contrast, the data for $x = 0.1$ are compatible with a $\sigma_{sc}$, that is purely originating from the second moment of the field distribution of vortex lattice. At low fields a maximum in $\sigma_{sc}(H)$ is observed followed by a decrease of the relaxation rate up to the highest fields. At first glance this appears to be consistent with a BCS $s$-wave superconductor. However, a quantitative analysis taking into account the large critical fields reveals strong discrepancies.

In a conventional $s$-wave superconductor the penetration depth $\lambda$ is field independent and $\sigma_{sc}$ increases with in-

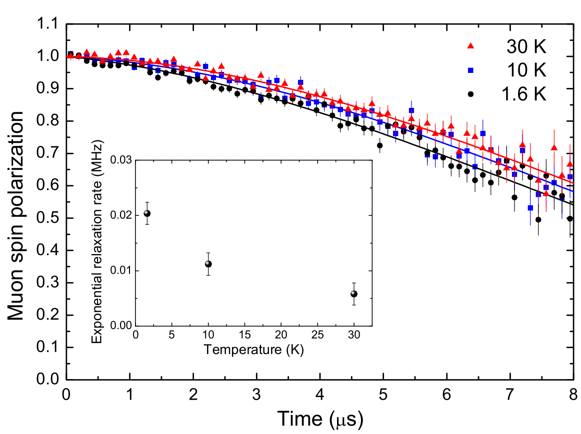


FIG. 2 (color online). Zero field $\mu$SR spectra of $LaFeAsO_{0.9}F_{0.1}$ for 1.6, 10, and 30 K. The very small temperature dependence of the ZF spectra is modeled by a multiplication of the Gaussian Kubo-Toyabe function with an exponential relaxation (the rate is shown in the inset).

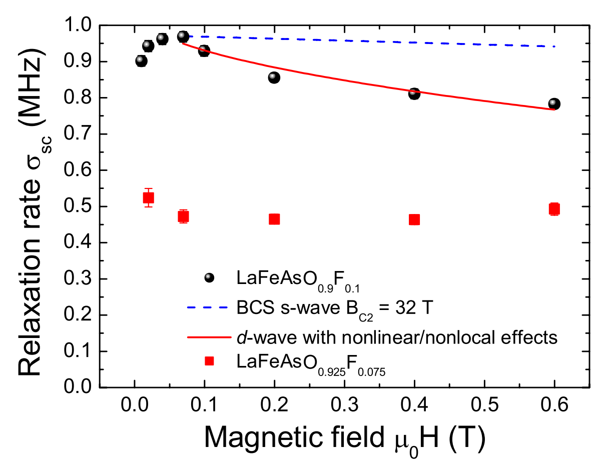


FIG. 3 (color online). Field dependence $\sigma_{sc}$ at 1.6 K for $LaFeAsO_{0.925}F_{0.075}$ and $LaFeAsO_{0.9}F_{0.1}$. The dashed line is the expected behavior for an $s$-wave BCS superconductor with $\mu_0 H_{c2} = 32$ T according to Eq. (1). The solid line is a fit of the data with Eq. (2) indicative for nodes in the gap function.

creasing magnetic field up to $H \simeq 2H_{c1}$. At higher fields a weak field dependence according to Eq. (1) is expected in an ideal triangular vortex lattice [23]:

$$\sigma_{sc}\,[\mu s^{-1}] = 4.83 \times 10^4(1-h)[1 + 3.9(1-h)^2]^{1/2}\lambda^{-2}\,[\mathrm{nm}]. \qquad (1)$$

Here, $h = H/H_{c2}$ and $H_{c2}$ is the upper critical field which has been reported to be as large as 32–65 T in optimally doped $LaFeAsO_{1-x}F_x$ [8,21,26]. We note that high field studies on our sample corroborate these large values [27]. Taking these critical fields, the theoretical BCS behavior has been calculated. It is shown by the dashed line in Fig. 3. The decrease of $\sigma_{sc}$ at $\mu_0 H \gtrsim 0.1$ T is in striking contradiction with conventional $s$-wave BCS behavior as given by Eq. (1). It is only reproduced if an unrealistically small $\mu_0 H_{c2} = 5$ T is assumed.

For unconventional superconductors with nodes in the gap, $\lambda$ depends on the field. This leads to a decrease of $\sigma_{sc}(H)$ for $H > 2H_{c1}$, which is generally observed for various high-$T_c$ superconductors [28]. This observation is nicely described by theories taking nonlocal or nonlinear effects into account. In the nonlinear case, the field dependence of the superfluid density can be described by [29,30]

$$\frac{\lambda^{-2}(H)}{\lambda^{-2}(H=0)} = \frac{\sigma_{sc}(H)}{\sigma_{sc}(H=0)} = 1 - K\sqrt{H}, \qquad (2)$$

where $K$ is the parameter depending on the strength of the effects. Applying this model, our data are well described by Eq. (2) with $K = 0.0034\ \mathrm{Oe}^{-0.5}$ indicative for superconductivity with nodes in the gap function [31].

It is well established that nodes in the gap function also influence the temperature dependence of the superfluid density $n_s/m^* \propto 1/\lambda^2$. This temperature dependence $n_s(T)$ is directly obtained from the temperature dependent TF-$\mu$SR measurements. To ensure an accurate determination of $\lambda(T)$ it is mandatory to measure slightly above the maximum of $\sigma_{sc}(H)$ where Eq. (1) is valid to determine the absolute value of $\lambda$. Therefore, the measurements were done with an external field of 700 G. The results for $\sigma_{sc}(T)$ are shown in Fig. 4.

Surprisingly, and in contrast to our conclusions from the field dependence, the results follow the $s$-wave weak coupling BCS temperature dependence as shown by the dashed curve. However, the data are also reasonably well described by a power law $1 - (T/T_c)^{2.45(4)}$, which is similar to the prediction for the dirty-limit $d$-wave model $1 - (T/T_c)^2$ [32]. Other gap symmetries such as a clean-limit $d$ wave or a nonmonotonic $d$ wave have been tested also, but were found to be inconsistent with the data. In particular, it is difficult to account for the very weak temperature dependence of $\sigma_{sc}$ at low $T$ within such models. Considering both field and temperature dependence of $\sigma_{sc}$, the dirty-limit $d$-wave model appears to be most compatible with the data. It is important to note that we observe a strong

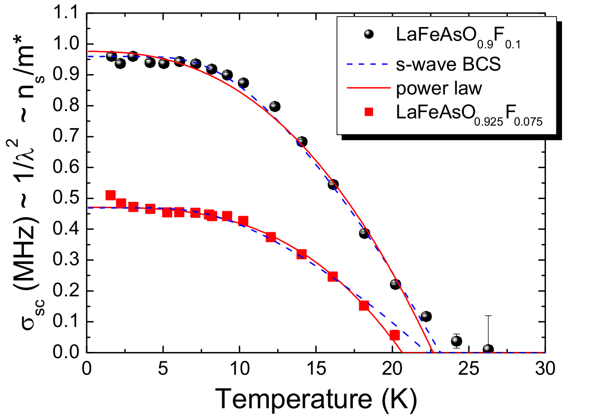


FIG. 4 (color online). Temperature dependence of $\sigma_{sc}$ measured in a field of $\mu_0 H = 0.07$ T for $LaFeAsO_{0.925}F_{0.075}$ and $LaFeAsO_{0.9}F_{0.1}$. The $\sigma_{sc}(T)$ were fitted using a standard BCS curve and a power law $1 - (T/T_c)^n$. In case of $LaFeAsO_{0.925}F_{0.075}$ the points below 4 K were omitted in the fit.

reduction of the high-TF Knight shift below $T_c$. This points to significant reduction of the spin susceptibility in the superconducting state which excludes triplet pairing.

The temperature dependence for the underdoped compound $LaFeAsO_{0.925}F_{0.075}$ resembles that of the optimum doped sample, with the difference that an upturn of $\sigma_{sc}$ is observed below 4 K. When measuring $\sigma_{sc}(T)$ in a higher field of 0.6 T, the upturn of $\sigma_{sc}(T)$ is already observed at higher temperatures $T \lesssim 7$ K. In other words, the upturn is not due to the temperature dependence of the superfluid density but a further hint to the proximity to magnetism which we have discussed in the context of the field dependent measurements shown in Fig. 3.

We now turn our discussion to $\lambda(T=0)$ and the in-plane penetration depth $\lambda_{ab}$. For $LaFeAsO_{0.9}F_{0.1}$ we determine a zero temperature relaxation rate $\sigma_{sc}(0) = 0.965(10)$ MHz. Using Eq. (1) the effective penetration depth can be calculated to $\lambda(0) = 333(2)$ nm. For $LaFeAsO_{0.925}F_{0.075}$ we analogously determined the low temperature relaxation rate to $\sigma_{sc}(0) = 0.47(2)$ MHz, yielding a penetration depth of $\lambda(0) = 477(10)$ nm. For powdered samples the experimentally extracted $\lambda$ is a geometrically averaged penetration depth. However, in $LaFeAsO_{1-x}F_x$ a rather large anisotropy can be expected from recent band structure calculations predicting, e.g., a resistivity ratio of $\rho_c/\rho_{ab} \approx 10-15$ [33]. In our TF-$\mu$SR measurements a large anisotropy of $\lambda$ is confirmed by the observation of a symmetric Gaussian shape of the field distribution $p(B)$. In contrast, for an isotropic superconductor a typical nonsymmetric shape of $p(B)$ with a van Hove singularity is found [28]. It has been shown [34] that for large anisotropies the measured effective penetration depth $\lambda$ becomes independent of the actual anisotropy and is solely determined by the in-plane penetration depth $\lambda_{ab}$ and can be expressed by $\lambda = 1.31\lambda_{ab}$. Therefore we obtain the in-plane penetration depth of

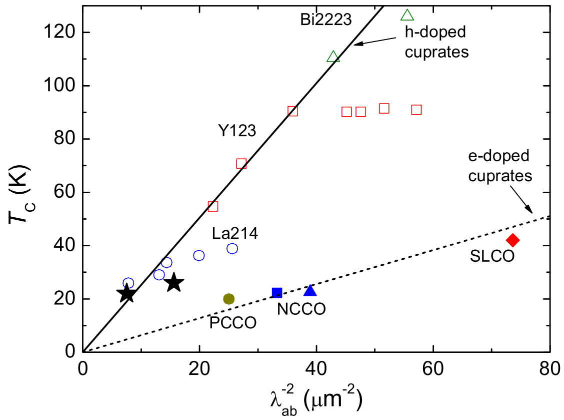


FIG. 5 (color online). Uemura plot for hole and electron doped high $T_c$ cuprates. Points for the cuprates are taken from [38]. The stars show the data for $LaFeAsO_{1-x}F_x$ obtained in this work.

$\lambda_{ab}(0) = 254(2)$ nm for $LaFeAsO_{0.9}F_{0.1}$ and $\lambda_{ab}(0) = 364(8)$ nm for $LaFeAsO_{0.925}F_{0.075}$, respectively.

In Fig. 5 we display our data in the so-called Uemura plot, which nicely demonstrates the linear relation of superfluid density and $T_c$ for underdoped and optimally doped superconductors [35]. We compare our data to the cuprate family. The data for $LaFeAsO_{1-x}F_x$ are close to the Uemura line for hole doped cuprates, indicating that the superfluid is also very dilute in the oxypnictides. This observation is in accordance with its small normal state charge carrier density [21,36,37] and provides further evidence for the unconventional superconductivity in $LaFeAsO_{1-x}F_x$.

In conclusion, we have performed zero field and high transverse field muon spin relaxation measurements on polycrystalline samples of $LaFeAsO_{1-x}F_x$ with $x = 0.075$ and 0.10. Our ZF-$\mu$SR experiments show that the spin-density state is completely suppressed upon $F$ doping, which suggests a close proximity of the superconducting phase to a magnetic quantum critical point. TF-$\mu$SR measurements reveal a weak temperature dependence of the superfluid density below $T_c/3$, which is consistent with both an $s$-wave and a dirty $d$-wave scenario. However, the field dependence supports an unconventional order parameter, but a clean-limit $d$ wave as well as triplet pairing can be clearly excluded by our data. We provide quantitative measurements of the in-plane penetration depths, which amount to $\lambda_{ab}(0) = 254(2)$ nm and $\lambda_{ab}(0) = 364(8)$ nm for $x = 0.1$ and $x = 0.075$, respectively. Hence, the superfluid in $LaFeAsO_{1-x}F_x$ is dilute and, remarkably, the data for the Fe-based superconductors are very close to the Uemura line of the hole doped high-$T_c$ cuprates.

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[1] Y. Kamihara *et al.*, J. Am. Chem. Soc. **130**, 3296 (2008).
[2] Z.-A. Ren *et al.*, Chin. Phys. Lett. **25**, 2215 (2008).
[3] C. Cao *et al.*, Phys. Rev. B **77**, 220506(R) (2008).
[4] H. Eschrig, arXiv:0804.0186.
[5] L. Shan *et al.*, arXiv:0803.2405.
[6] G. Mu *et al.*, Chin. Phys. Lett. **25**, 2221 (2008).
[7] C. Ren *et al.*, arXiv:0804.1726.
[8] F. Hunte *et al.*, Nature (London) **453**, 903 (2008).
[9] G. Xu *et al.*, Europhys. Lett. **82**, 67 002 (2008).
[10] I. Mazin *et al.*, Phys. Rev. Lett. **101**, 057003 (2008).
[11] K. Kuroki *et al.*, arXiv:0803.3325 [Phys. Rev. Lett. (to be published)].
[12] X. Dai *et al.*, Phys. Rev. Lett. **101**, 057008 (2008).
[13] Q. Han *et al.*, arXiv:0803.4346.
[14] L. Boeri *et al.*, Phys. Rev. Lett. **101**, 026403 (2008).
[15] P. A. Lee and X. G. Wen, arXiv:0804.1739.
[16] F. Ma *et al.*, Phys. Rev. B **78**, 033111 (2008).
[17] J. Dong *et al.*, Europhys. Lett. **83**, 27 006 (2008).
[18] C. Cruz *et al.*, Nature (London) **453**, 899 (2008).
[19] A. M. McGuire *et al.*, arXiv:0804.0796.
[20] G. Giovannetti *et al.*, arXiv:0804.0866.
[21] X. Zhu *et al.*, Supercond. Sci. Technol. **21**, 105001 (2008).
[22] R. S. Hayano *et al.*, Phys. Rev. B **20**, 850 (1979).
[23] E. H. Brandt, Phys. Rev. B **37**, 2349 (1988).
[24] A. T. Savici *et al.*, Phys. Rev. Lett. **95**, 157001 (2005).
[25] R. Khasanov *et al.*, Phys. Rev. B **76**, 094505 (2007).
[26] A. S. Sefat *et al.*, Phys. Rev. B **77**, 174503 (2008).
[27] G. Fuchs *et al.* (to be published).
[28] J. E. Sonier, J. H. Brewer, and R. F. Kiefl, Rev. Mod. Phys. **72**, 769 (2000).
[29] H. Won and K. Maki, Europhys. Lett. **54**, 248 (2001).
[30] I. Vekhter, J. P. Carbotte, and E. J. Nicol, Phys. Rev. B **59**, 1417 (1999).
[31] We note that a maximum in $\sigma_{sc}(H)$ may be attributed to vortex lattice distortions caused by strong pinning. However, in our $x = 0.1$ sample pinning is weak as signaled by the field cooled susceptibility shown in Fig. 1.
[32] P. J. Hirschfeld, W. O. Putikka, and D. J. Scalapino, Phys. Rev. B **50**, 10 250 (1994).
[33] D. J. Singh and M. H. Du, Phys. Rev. Lett. **100**, 237003 (2008).
[34] V. I. Fesenko, V. N. Gorbunov, and V. P. Smilga, Physica (Amsterdam) **176C**, 551 (1991).
[35] Y. J. Uemura *et al.*, Phys. Rev. Lett. **62**, 2317 (1989).
[36] G. F. Chen *et al.*, Phys. Rev. Lett. **101**, 057007 (2008).
[37] H. Yang *et al.*, arXiv:0803.0623.
[38] Y. J. Uemura *et al.*, Phys. Rev. Lett. **66**, 2665 (1991); A. Shengelaya *et al.*, Phys. Rev. Lett. **94**, 127001 (2005); A. A. Nugroho *et al.*, Phys. Rev. B **60**, 15 384 (1999); C. C. Homes *et al.*, Phys. Rev. B **56**, 5525 (1997); C. C. Homes *et al.*, Phys. Rev. B **74**, 214515 (2006).