Muon-spin rotation study of the in-plane magnetic penetration depth of FeSe_{0.85}: evidence for nodeless superconductivity

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The recent discovery of the Fe-based high-temperature superconductors has attracted considerable attention to the pnictides. The superconductivity was first found in LaO_{1-x}F_xFeAs [1] and, later on, in other single-layer arseno-pnictides by replacing La with various rare-earth elements (Sm, Nd, Pr, Ce, Ho, Y, Dy, and Tb) [2] as well as in oxygen-free compounds such as doped double-layer FeSe [3] and single-layer LiFeAs [4]. The common structural feature of all these materials is the Fe-layers consisting of an Fe square planar sheet tetrahedrally coordinated by As. Recently, superconductivity with the transition temperature \( T_c \approx 8 \) K was discovered in \( \alpha-FeSe \) with PbO-structure [5]. This compound also has a Fe square lattice with Fe atoms tetrahedrally coordinated by Se ones, similar to the structure of FeAs planes in the single- and the double-layer arseno-pnictides. In this respect FeSe, consisting of the "superconducting" Fe-Se layers only, can be treated as a prototype of the known families of FeAs-based high-temperature superconductors and, consequently, becomes a good modeling system to study the mechanism leading to the occurrence of superconductivity in these new class of materials.

Here we report a study of the in-plane magnetic field penetration depth \( \lambda_{ab} \) in iron selenide superconductor with the nominal composition FeSe_{0.85} by means of muon-spin rotation (\( \mu \)SR). \( \lambda_{ab}^{2}(T) \) was reconstructed from the temperature dependences of the \( \mu \)SR linewidth measured in a magnetic field of 0.01 T. The observed \( \lambda_{ab}^{2}(T) \) was found to be equally well described within the framework of anisotropic \( s-\)wave as well two-gap \( s + s-\)wave models. In a case of anisotropic \( s-\)wave model the maximum value of the gap at \( T = 0 \) was found to be \( \Delta_0 = 1.35 \) meV leading to \( 2\Delta_0/k_BT_c = 3.79 \), close to the weak-coupling BCS value 3.52. The two-gap \( s + s-\)wave model yields \( \Delta_{0,1} = 1.63 \) meV and \( \Delta_{0,2} = 0.38 \) meV. The corresponding gap to \( T_c \) ratios are \( 2\Delta_{0,1}/k_BT_c = 4.59 \) and \( 2\Delta_{0,2}/k_BT_c = 1.07 \) close to those reported for various single- and double-layer arseno-pnictide superconductors [6, 7, 8].

Details on the sample preparation for FeSe_{0.85} can be found elsewhere [9]. X-ray diffraction analysis reveals that the \( \alpha-FeSe \) phase is dominant and that the amount of the impurity fraction does not exceed \( \approx 7\)-10%. The AC magnetization (\( M_{AC} \)) measurements (\( \mu_0H_{AC} = 0.1 \) mT, \( \nu = 1000 \) Hz) were performed on a Quantum Design PPMS magnetometer at temperatures ranging from 1.75 K to 300 K. The superconducting transition temperature \( T_c = 8.26(2) \) K was obtained as an intersection of the linearly extrapolated \( M_{AC}(T) \) with \( M_{AC} = \text{const} \) line (see Fig. 1).

FIG. 1: (Color online) Temperature dependence of the AC magnetization \( M_{AC} (\mu_0H_{AC} = 0.1 \) mT, \( \nu = 1000 \) Hz) of FeSe_{0.85}.

Zero field (ZF), longitudinal field (LF) and transverse field (TF) \( \mu \)SR experiments were performed at the
\[ A^{ZF}(t) = A_0 \exp(-\Delta t). \]  

Here \( A_0 \) is the initial asymmetry at \( t = 0 \) and \( \Delta \) is the exponential depolarization rate. The results of the analysis of the ZF data and the representative ZF and LF muon-time spectra are shown in Fig. 2. The open and the closed symbols are from the measurements taken on the LTF and the GPS instruments, respectively.

The exponential character of the muon polarization decay might be explained either by existence of fast electronic fluctuations measurable within the \( \mu \)SR time-window [4] or by a static magnetic field distribution caused by diluted and randomly oriented magnetic moments [10]. To distinguish between these two cases LF \( \mu \)SR experiments were performed. As is shown in Ref. [14] in a case when the applied longitudinal field is much stronger than the internal field (\( B > 10 B_{int} \)) the muon spins become "decoupled" from the static internal field. On the other hand, field fluctuations perpendicular to the applied external field can cause irreversible spin-flip transitions of the muon spin, leading to depolarization [11]. As is shown in the inset of Fig. 2 an external field of 0.01 T is already enough to completely decouple the muon spins.

This proves that the magnetism in FeSe\(_{0.85}\) sample studied here is static in origin and is caused by diluted and randomly distributed magnetic moments. Bearing this in mind and by taking into account the presence of the relatively high paramagnetic contribution at \( T > T_c \) (see Fig. 1) we may conclude that the magnetism observed in both, ZF \( \mu \)SR and magnetization experiments, has similar sources and, most probably, caused by the traces of Fe impurities [5].

**FIG. 2:** (Color online) Temperature dependence of the ZF muon depolarization rate \( \Delta \) of FeSe\(_{0.85}\). The inset shows ZF (\( T = 1.5 \) K and 10 K) and LF (\( T = 10 \) K, \( \mu_0 H = 0.01 \) T) \( \mu \)SR time-spectra of FeSe\(_{0.85}\).

**FIG. 3:** (Color online) Temperature dependence of the Gaussian depolarization rate \( \sigma \) at \( \mu_0 H = 0.01 \) T of FeSe\(_{0.85}\). The inset shows the TF muon-time spectra above (\( T = 10 \) K) and below (\( T = 1.5 \) K) the superconducting transition temperature \( T_c = 8.26 \) K.

The in-plane magnetic penetration depth \( \lambda_{ab} \) was studied in the TF \( \mu \)SR experiments. In a powder sample the magnetic penetration depth \( \lambda \) can be extracted from the Gaussian muon-spin depolarization rate \( \sigma_{sc}(T) \propto 1/\lambda^2(T) \), which probes the second moment of the magnetic field distribution in the mixed state [12]. For highly anisotropic layered superconductors (like the pnictide superconductors) \( \lambda \) is mainly determined by the in-plane penetration depth \( \lambda_{ab} \) [13]: \( \sigma_{sc}(T) \propto 1/\lambda_{ab}^2(T) \). By taking into account the weak magnetism observed in our ZF experiments (see Fig. 2 and discussion above) the TF \( \mu \)SR data were analyzed by using the following functional form:

\[ A^{TF}(t) = A_0 \exp(-\Delta t) \exp(-\sigma^2 t^2/2) \cos(2\gamma B_{int}t + \phi). \]  

Here \( \gamma/2\pi = 135.5 \text{ MHz/T} \) is the muon gyromagnetic ratio, \( \phi \) is the initial phase of the muon-spin ensemble, and \( \sigma = (\sigma^2_{sc} + \sigma^2_{nm})^{0.5} \) is the Gaussian relaxation rate. \( \sigma_{nm} \) is the nuclear magnetic dipolar contribution which is generally temperature independent [14]. The whole set of 0.01 T TF \( \mu \)SR data was fitted simultaneously with \( A_0, \Delta, \) and \( \phi \) as a common parameters and \( \sigma \) and \( B_{int} \) as individual parameters for each temperature point. The exponential relaxation rate was assumed to be temperature independent in accordance with the results of our
ZF μSR experiments (see Fig. 2). $\sigma_{nm}$ was fixed to the value obtained above $T_c$ where $\sigma = \sigma_{nm}$ (see Fig. 3). The results of the analysis and the representative TF muon-time spectra are shown in Fig. 3. The open and the closed symbols are again from the measurements taken on the LTF and the GPS instruments, respectively.

The superconducting part of the Gaussian depolarization rate $\sigma_{sc}$ can be converted into $\lambda_{ab}$ via 9,12,13:

$$\sigma_{sc}^2/\lambda_{sc}^2 = 0.00126 \Phi_0^2/\lambda_{ab}^4,$$

where $\Phi_0 = 2.068 \cdot 10^{-15}$ Wb is the magnetic flux quantum. Fig. 4 shows $\lambda_{ab}^{-2}(T)$ obtained from the measured $\sigma_{ar}(T)$ by means of Eq. 3. Regarding the pairing symmetry, available experimental results on various single- and double-layer arseno-pnictides are divided between those favoring an isotropic 16,17 as well as an anisotropic 18 nodeless gap and those supporting line nodes 19. The two-gap behavior was also reported in Refs. 3,8,16,20,21. Bearing this in mind the data in Fig. 4 were analyzed by using single-gap and two-gap models, assuming that the superconducting energy gaps have the following symmetries: $s$–wave (a), $s+s$–wave (b), anisotropic $s$–wave (c), and $d$–wave (d).

Temperature dependence of the magnetic penetration depth $\lambda$ was calculated within the local (London) approximation ($\lambda \gg \xi$) by using the following functional form 22,23:

$$\lambda^{-2}(T) = 1 + \frac{1}{\pi} \int_0^{2\pi} \int_{\Delta(T,\varphi)}^{\infty} \left( \frac{\partial f}{\partial E} \right) \frac{E dE d\varphi}{\sqrt{E^2 - \Delta(T,\varphi)^2}}.$$  

Here $\lambda^{-2}(0)$ is the zero-temperature value of the magnetic penetration depth, $f = [1 + \exp(E/k_B T)]^{-1}$ is the Fermi function, $\varphi$ is the angle along the Fermi surface, and $\Delta(T,\varphi) = \Delta_0 \delta(T/T_c) g(\varphi)$ ($\Delta_0$ is the maximum gap value at $T = 0$). The temperature dependence of the gap is approximated by $\delta(T/T_c) = \tanh\{1.82[1.018(T_c/T - 1)^{0.51}]\}$ 24. The function $g(\varphi)$ describes the angular dependence of the gap and is given by $g^s(\varphi) = 1$ for the $s$–wave gap, $g^d(\varphi) = |\cos(2\varphi)|$ for the $d$–wave gap, and $g^{s\Delta N}(\varphi) = (1 + a \cos 4\varphi)/(1 + a)$ for the anisotropic $s$–wave gap 23.

The two-gap calculations were performed within the framework of the so called $\alpha$–model assuming that the total superfluid density is a sum of two components 22,24:

$$\lambda^{-2}(T) = \omega \cdot \lambda^{-2}(T,\Delta_0,1) + (1 - \omega) \frac{\lambda^{-2}(T,\Delta_0,2)}{\lambda^{-2}(0,\Delta_0,2)}.$$

Here $\Delta_0,1$ and $\Delta_0,2$ are the zero-temperature values of the large and the small gap, respectively, and $\omega (0 \leq \omega \leq 1)$ is the weighting factor which represents the relative contribution of the larger gap to $\lambda^{-2}$.

The results of the analysis are presented in Fig. 4 by solid black lines. The angular dependences of the gaps are shown as the insets.
[\Delta_0 \cdot g(\varphi)] are shown in the corresponding insets. It is obvious that the simple s- and d-wave approaches cannot describe the observed \lambda_{ab}^2(T) [see Figs. 1 (a) and (d)]. In both cases the low temperature points stay systematically higher than the theoretically derived curves. The constant, within our experimental uncertainty, \lambda_{ab}^{-2}(T) at T \lesssim 1 K is also inconsistent with the presence of any type of nodes in the energy gap of FeSe_{0.85}. In contrast, both, anisotropic s- and two-gap s + s-wave models [Figs. 1(b), (c)] describe the experimental data reasonably well. In the following we are going to discuss separately the results obtained within the framework of these two models. For the anisotropic s-wave case we get \Delta_0 = 1.35 meV, a = 0.796, and \lambda_{ab}(0) = 406 nm. The corresponding gap to \Tc ratio is 2\Delta/k_BTc = 3.79 which is rather close to the weak-coupling BCS value 3.52. The obtained variation with angle \Delta_{max}/\Delta_{min} = 1.35/0.153 \approx 8.8 is substantially bigger than 1.2 reported in Ref. 18 for NdFeAsO_9F_{0.1}. The fit within the two-gap s + s-wave model yields \Delta_{0,1} = 1.63 meV, \Delta_{0,2} = 0.38 meV, \omega = 0.654, and \lambda_{ab}(0) = 403 nm. It is interesting to note that the "large" and the "small" gap to \Tc ratios 2\Delta_{0,1}/k_BTc = 4.59 and 2\Delta_{0,2}/k_BTc = 1.07 are very close to those reported for various single- and double-layer arsene-antimonite superconductors based on the results of the point contact Andreev reflection spectroscopy experiments of Szabo et al. 6 and Gonelli et al. 7, and the first critical field measurements of Ren et al. 8. The multiple gaps may originate from the multiple bands at the Fermi level of FeSe_{0.85}. First-principle calculation indicates that the Fermi surface (FS) of FeSe is quasi-two dimensional and consists of hole-type sheets around the \Gamma point and electron-type sheets around the M point of the Brillouin zone 20. It is conceivable that the two bands open up on the different sheets of the FS. In this context, the present compound may resemble the situation of MgB2 where a large gap opens on the FS derived from the orbitals in the boron plane, while a small gap opens on the FS derived from orbitals perpendicular to the boron plane 27.

To conclude, muon-spin rotation measurements were performed on the superconductor FeSe_{0.85} (\Tc \approx 8.3 K). \lambda_{ab}^2(T) was reconstructed from the temperature dependence of the Gaussian muon depolarization rate measured at \mu_0H = 0.01 T. The absolute value of the in-plane magnetic penetration depth \lambda_{ab} at T = 0 was estimated to be \lambda_{ab}(0) \approx 405 nm. The temperature dependence of \lambda_{ab}^{-2} was found to be inconsistent with an isotropic s-wave as well as with a d-wave symmetry of the superconducting energy gap. A good agreement between the experimental data and the theory was obtained within the framework of a two-gap s + s and an anisotropic s-wave gap models thus suggesting that the superconducting energy gap in FeSe_{0.85} superconductor is fully developed and contains no nodes.

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