Direct evidence of superconductivity and determination of the superfluid density in buried ultrathin FeSe grown on SrTiO$_3$

P. K. Biswas,$^1$ Z. Salman,$^1$ Q. Song,$^2$ R. Peng,$^2$ J. Zhang,$^2$ L. Shu,$^2$ D. L. Feng,$^2$ T. Prokscha,$^1$ and E. Morenzoni$^1$

$^1$Laboratory for Muon Spin Spectroscopy, Paul Scherrer Institut, CH-5232 Villigen PSI, Switzerland
$^2$State Key Laboratory of Surface Physics, Department of Physics, Fudan University, Shanghai 200433, China

(Dated: January 16, 2018)

Bulk FeSe is superconducting with a critical temperature $T_c \approx 8$ K and SrTiO$_3$ is insulating in nature, yet high-temperature superconductivity has been reported at the interface between a single-layer FeSe and SrTiO$_3$. Angle resolved photoemission spectroscopy and scanning tunneling microscopy measurements observe a gap opening at the Fermi surface below $\approx 60$ K. Elucidating the microscopic properties and understanding the pairing mechanism of single-layer FeSe is of utmost importance as it is a basic building block of iron-based superconductors. Here, we use the low-energy muon spin rotation/relaxation technique (LE-$\mu$SR) to detect and quantify the supercarrier density and determine the gap symmetry in FeSe grown on SrTiO$_3$ (100). Measurements in applied field show a temperature dependent broadening of the field distribution below $\sim 60$ K, reflecting the superconducting transition and formation of a vortex state. Zero field measurements rule out the presence of magnetism of static or fluctuating origin. From the inhomogeneous field distribution, we determine an effective sheet supercarrier density $n_s^{2D} \approx 6 \times 10^{14}$ cm$^{-2}$ at $T \rightarrow 0$ K, which is a factor of 4 larger than expected from ARPES measurements of the excess electron count per Fe of 1 monolayer (ML) FeSe. The temperature dependence of the superfluid density $n_s(T)$ can be well described down to $\sim 10$ K by simple s-wave BCS, indicating a rather clean superconducting phase with a gap of $10.2(1.1)$ meV. The result is a clear indication of the gradual formation of a two dimensional vortex lattice existing over the entire large FeSe/STO interface and provides unambiguous evidence for robust superconductivity below 60 K in ultrathin FeSe.

I. INTRODUCTION

Following the discovery of high-$T_c$ cuprates,$^{11,12}$ a few decades ago, the Fe-based superconductors,$^{3,15}$ represented an additional novel and important class of high-$T_c$ superconductors displaying, however, average critical temperatures lower than the cuprates. Surprisingly, high-temperature superconductivity with a $T_c \approx 60$-70 K was found in single-layer FeSe on SrTiO$_3$ (STO).$^{2,13}$ Similar high temperatures exceeding that of all known bulk iron-based superconductors have also been achieved on other oxide substrates.$^{13}$

This finding is extremely important in view of the simple crystal structure of the system, which consists of a single Se-Fe-Se unit, i.e. the basic building block of all iron-chalcogenide superconductors, and may pave the way to identifying key ingredients of high-$T_c$ superconductivity.$^{10}$ Single-layer FeSe exhibits a distinct electronic structure with only electron pockets near the Brillouin zone corner$^{11,13}$ This is in contrast to its bulk counterpart, which also shows hole pockets at the zone center.

Transport measurements performed ex situ find, with respect to bulk, an enhancement of $T_c$ with onset around 40 K not only in 1 ML FeSe$^{13}$ but also in ultrathin layers in various configurations$^{15}$ including electric-double-layer transistor films$^{13}$ and ultrathin flakes on SiO$_2$/Si$^{17}$. Similar $T_c$ as on STO have been measured on other substrate materials such as MgO, KTaO$_3$, TiO$_2$ (rutile$^{13}$ and anatase phase$^{13}$) and K-doped FeSe films$^{10}$ whereas in situ zero-resistivity was detected at a temperature as high as 109 K$^{21}$. Diamagnetic shielding was also observed up to $T_{\text{onset}} \sim 65$ K$^{22}$.

The superconducting gap of FeSe/STO has been mainly characterized by surface sensitive techniques such as ARPES and STM. The data suggest that single-layer FeSe has plain s-wave pairing symmetry.$^{10,11,13,22}$ However, on its own, detection of a gap appearing below $\sim 60$ K does not provide conclusive evidence that it is only related to the formation of a condensate of Cooper pairs and does not exclude other contributions such as magnetic, charge or spin density wave gaps. Transport measurements, on the other hand, cannot easily discriminate between filamentary and bulk superconductivity. It is therefore essential to characterize the presence of superconductivity in FeSe/STO and its microscopic properties by other techniques, providing complementary information such as the superfluid density and the homogeneity of the superconducting phase.

Here, we report detailed depth-resolved investigation of the superconducting and magnetic properties in ultra-thin FeSe by the low-energy muon spin rotation/relaxation (LE-$\mu$SR) technique. Zero field (ZF) $\mu$SR measurements demonstrate that the ground state is non-magnetic and transverse field (TF) $\mu$SR results show that superconductivity appears below 62 K. Taking into account the extreme 2D-character of the vortex state, we estimate the effective superfluid sheet density $n_s^{2D}(T)$.

Its temperature dependence is well described down to $\sim 10$ K by a simple BCS s-wave model, with a gap $\Delta(0) = 10.2(1.1)$ meV.
II. EXPERIMENTAL DETAILS

A. Film growth and characterization

Figure 1 shows a schematic of the heterostructure used in this experiment. Single-layers FeSe thin films were grown using molecular beam epitaxy (MBE) on a $10 \times 10 \text{ mm}^2$ TiO$_2$ terminated and Nb-doped (0.5% wt) (001)-oriented SrTiO$_3$ substrate. The substrate was pre-cleaned following the method described in previous work and ultrahigh vacuum (UHV) condition was maintained during deposition to enable continuous in situ growth. In the UHV chamber the substrate was degassed at 550°C for three hours and then heated to 950°C under a Se (99.9999%) flux for 30 minutes. It was kept at 490°C in Se and Fe (99.995%) flux for co-evaporation and co-deposition with the flux ratio of 20:1. After growth, the films were annealed at 600°C in vacuum for 3h. In-situ measurements confirmed the possible 60 K superconductivity in the monolayer (ML) FeSe film. Four more unit cells of FeSe thin films were successfully grown above the single-layer FeSe. The additional layers were deposited for stabilization purpose, since, surprisingly the original tunneling spectra of two unit cells or thicker FeSe films did not show signs of superconductivity. Before depositing the overlayers the FeSe ML was characterized by ARPES. Figure 2 shows the result exhibiting the typical features of the electronic structure (not to scale) of the heterostructure with a ultrathin FeSe film grown on the SrTiO$_3$ substrate. For transverse field measurements the magnetic field is applied perpendicular to the sample surface. The polarization of the implanted muons is parallel to the sample surface.

B. Low-energy $\mu$SR

To measure the local magnetic and superconducting properties of the ultrathin FeSe layer we use LE-$\mu$SR as a sensitive magnetic probe. Fully polarized muons are implanted in the sample one at a time, where they thermalize and act as sensitive magnetic microprobe. The muon spin precesses around the local magnetic field $B$ at the muon site with the Larmor frequency $\omega_\mu = \gamma_\mu B$, $\frac{2\pi}{\gamma_\mu} = 135.5 \text{ MHz/T}$. The precession and relaxation of the spin ensemble leads to a temporal evolution of the polarization, which is easily detectable via the asymmetric muon decay (lifetime $\tau_\mu = 2.2 \mu$s), where a positron is emitted preferentially in the direction of the muon spin at the moment of the decay. From the damped precession signal the field distribution associated with the vortex state can be determined. The LE-$\mu$SR experiments were performed on the LEM instrument, at the $\mu$E4 beamline of the Paul Scherrer Institut in Villigen, Switzerland. Here the energy of the muons can be tuned ($\sim 1$ to 30...
keV) to control the implantation depth in the range (∼1-300) nm and thus to probe the magnetic response in different layers of the heterostructure. With this unique ability, the LE-µSR technique is an ideal probe for studying the superconducting properties of the FeSe layer by implanting the muons on or very close to this layer. This procedure has been successfully applied to address related questions in a variety of systems and heterostructures. In particular, by varying the implantation energy of the muons, the spatial evolution of the magnetic field distribution as the flux lines emerge through the surface of a superconducting YBa$_2$Cu$_3$O$_{7-δ}$ film has been monitored, superconducting proximity effects of buried cuprate layers, the paramagnetic Meissner effect due to spin triplet component and magnetism at transition metal-molecular interfaces have been detected.

C. Zero-field and transverse-field µSR measurements

Initially, we tuned the muon beam implantation energy $E$ to maximize the fraction of muons stopping in the vicinity of the FeSe single-layer. Monte Carlo simulations, presented in Figure 3, show that this is achieved for $E ∼ 3$ keV. The program TRIM.SP, specially modified for muon implantation in heterostructures and whose reliability to calculate stopping profiles has been previously tested, was used for the calculation.

![Figure 3. Muon implantation profiles.](image)

We performed ZF and TF-µSR measurements at different temperatures. A ZF measurement is very sensitive to magnetism; in a magnetic environment, well defined precession frequencies may be observed in the case of long-range order. Alternatively a distribution of precession frequencies with the corresponding width proportional to the field inhomogeneity may be detected. If the field distribution is broad when averaged over the sample, as in the case of disordered or short range magnetism, the muon decay asymmetry displays a fast depolarization. In the case of dynamic moments with fluctuating times within the µSR time window, spin relaxation is also observed. These features allow the direct observation of the onset of magnetic order even if very weak. It has been used for instance to search for time-reversal symmetry breaking phenomena in the superconducting phase, where a very tiny spontaneous static magnetic field appears with the onset of superconductivity. The ZF-

![Figure 4. ZF muon spin relaxation.](image)

spectra taken at 2.3 keV muon implantation energy can be described well using a static Gaussian Kubo-Toyabe relaxation function, where the time evolution of the asymmetry $A(t)$, which is proportional to the muon spin polarization, is given by:

$$A(t) = A_0 \left\{ \frac{1}{3} + \frac{2}{3} \left( 1 - \sigma_{ZF}^2 t^2 \right) \exp \left( -\frac{\sigma_{ZF}^2 t^2}{2} \right) \right\},$$

where $A_0$ is the initial asymmetry and $\sigma_{ZF}$ the muon spin relaxation rate. We do not detect any difference in the spectra, taken at 5 K and 100 K, as shown in Figure 4. The nearly equal and very small values of $\sigma_{ZF}$ (0.086(5) and 0.082(5) $\mu$s$^{-1}$ for 5 and 100 K, respectively), extracted from the fits for two different temperatures, reflect the presence of random local magnetic fields arising solely from the nuclear moments in the sample.

For the TF-µSR measurements as a function of temperature, the sample was cooled in a magnetic field of 10 mT applied normal to the sample surface and to the initial muon spin direction.

Figure 5 shows the TF-µSR time spectra collected at (a) 5 K and (b) 70 K. At 70 K, the local field probed...
III. RESULTS AND DISCUSSION

A. Temperature and energy dependence of the field broadening

The temperature dependence of the Gaussian damping rate $\sigma(T) = (\sigma_{\text{sc}}^2(T) + \sigma_{\text{nm}}^2)^{\frac{1}{2}}$ is shown in Figure 6. The data displays a clear increase of $\sigma$ with lowering the temperature due to the term $\sigma_{\text{sc}}(T) = \gamma_\mu \sqrt{\Delta B^2}$, which expresses the inhomogeneous field distribution associated with the formation of the vortex state in superconducting FeSe below $\sim 60$ K. $\sigma_{\text{nm}}$ ($\approx \sigma_{\text{ZF}}$) is caused by the dipolar field contribution of the nuclear moments and is temperature independent. The average spin precession frequency, which is proportional to the average local field, corresponds very closely to the applied field as expected from a demagnetizing factor close to one in our geometry. Our $\textit{ex situ}$ value of $T_c$ agrees well with the temperature for gap opening observed in several $\textit{in situ}$ ARPES measurements.

The measurement at 5 K of $\sigma$ as a function of depth by varying the muon implantation energy, $E$, further establishes the source of the observed superconductivity. As expected from the TRIM.SP calculations, we observe the largest field inhomogeneity at $\sim 3$ keV, where most of the muons are implanted very close to the FeSe layers. $\sigma_{\text{nm}}$ is small and temperature independent but slightly depends on the muon implantation energy due to the different nuclear moment contribution in the various layers composing the heterostructure. We determined this contribution by performing a full energy scan in the normal state at $T = 100$K and corrected for it to obtain the energy dependence of the field broadening $\sigma_{\text{sc}}$ in the vortex state of FeSe (Fig. 7).

\[
A(t) = A_0 \exp \left(-\sigma^2 t^2 / 2\right) \cos(\gamma_\mu B t + \phi), \quad (2)
\]

where $A(0)$ is the initial asymmetry, $B$ is the magnetic field at the muon sites, $\phi$ is the initial phase of the muon polarization precession signal, and $\sigma(T)$ is the spin damping rate due to the field inhomogeneities.
We determine $B_z(x, y, z)$ from the requirement that it fulfills London equation with source terms representing the flux lines core in a very thin superconducting film ($-d/2 < z < d/2$) and Laplace equation outside

$$-\nabla^2 B_z(x, y, z) + \Pi(z) \frac{B_z(x, y, z)}{\lambda^2} = \Pi(z) \frac{\Phi_0}{\lambda^2} \sum_R \delta(\vec{r} - \vec{R})$$

(3)

where $\Pi(z)$ is the boxcar function, which is equal to 1 for $-d/2 \leq z \leq d/2$ and 0 otherwise, $\vec{r} = (x, y)$ and $\vec{R}$ the vortex positions. The solution is obtained by decomposing $B_z(x, y, z)$ into its Fourier components in the $x - y$ plane

$$B_z(x, y, z) = \sum_{k \neq 0} b_z(\vec{k}, z) e^{-i\vec{k} \cdot \vec{r}}$$

(4)

where $\vec{k}$ is the reciprocal lattice vector of the flux lattice with $k = |\vec{k}| = \sqrt{\frac{16\pi^2(m^2+n^2)}{\gamma^2\lambda^2}}$, $m, n$ integer. After matching the field and its derivative at the layer boundaries, we determine the Fourier coefficients $b_z(\vec{k}, z)$ so that solutions are obtained inside and outside the single-layer FeSe. The width of the field distribution at $z$ is then given by $\Delta B_z^2(z) = \langle B_z^2(z) \rangle - \langle B_z(z) \rangle^2 = \sum_{k \neq 0} b_z(k, z)^2$. Averaging is over the $x$ and $y$ plane coordinates. For a comparison with the measured broadening, $\Delta B_z(z)$ has to be weighted with the normalized muon stopping distribution at $n(z, E)$ so that $\sigma_{sc}(E) = \gamma^2 \frac{\int_{-\infty}^{\infty} \Delta B_z^2(z)n(z, E)dz}{\int_{-\infty}^{\infty} n(z, E)dz}$. In contrast to the 3D case where $\sigma_{sc} \propto \frac{1}{\xi}$, in our 2D situation we find that the field broadening is governed by the Pearl length scale $\Delta \nu = 2\lambda^2/d$ as expected for the vortex state in superconducting films with $d < \lambda$. For instance, taking into account that the superconducting layer is very thin and that the dominating contribution to the observed field broadening comes from the muons stopping outside the layer ($d/2 < z < -d/2$), one finds that the Fourier coefficients can be expressed as $b_z(k, z) \approx 2\gamma \sqrt{\frac{2m\nu}{\lambda^2}} e^{-\left(kz_{eff}\right)}$.

C. Determination of microscopic superconducting properties

The Pearl length scale is directly related to the sheet superconducting carrier density $n_{s}^{2D} = 2m\nu/\lambda^2$. Figure 8 shows the temperature dependence of the sheet superfluid density in the ultrathin FeSe layer. Remarkably, $n_{s}^{2D}$ does not show any signs of phase fluctuations, which may be expected in a 2D-like superconductor, probably because of the strong coupling to the STO substrate. This temperature dependence can be well fitted down to 10 K using a single-gap BCS $s$-wave model (solid line in Figure 8). The fit gives a gap value at zero temperature $\Delta(0) = 10.2(1.1)$ meV and $T_c = 62(2)$ K. This

B. Calculation of the field width

$\mu$SR has been widely used to characterize the properties of bulk superconductors and determine their microscopic parameter. For a bulk superconductor in the vortex state the field broadening is directly given by the magnetic penetration depth $\sigma_{sc} \propto \frac{1}{\sqrt{\lambda}}$. In our sample $\sigma_{sc}(T)$ is determined by the 2D pancake-like vortices that form in a thin superconducting layer. Since the muon stopping profile encompasses a region outside the single FeSe layer (see Figure 3), the inhomogeneous stray field of the vortices, which extends outside the superconducting layer, has to be taken into account to obtain the relationship between $\sigma_{sc}(T)$ and the effective superfluid density in FeSe.

The field profile and distribution have been obtained by solving the London equation, which is appropriate for an extreme type-II superconductor ($\xi \ll \lambda$, $\xi$ coherence length $\sim 2-3$ nm). For the ultrathin FeSe layers application of a magnetic field will lead to the formation of a regular vortex structure of hexagonal symmetry, with each vortex carrying a flux quantum $\Phi_0$ and intervortex separation $D \equiv \sqrt{\frac{2 \Phi_0}{\sqrt{3} \mu_0 B_0}} \approx 490$ nm for $B_0 = 10$ mT. Indication of such a structure has been visualized by STM measurement. In a bulk superconductor the local field $B_z(x, y, z)$, although varying with the planar coordinates $x$ and $y$, is always parallel to the applied field and perpendicular to the sample surface ($z$ direction, $z = 0$ center of the single layer). In our case, near the single-layer, the field lines splay out. However, this effect on the $\mu$SR signal is small and we can consider the normal component of the field.
The solid curve is a fit with a BCS s-wave gap. For comparison a model assuming an additional small gap manifesting itself at low temperatures is shown as a dashed line.

FIG. 8. Temperature dependence of the superfluid sheet density. Superfluid 2D density versus temperature for ultrathin FeSe. The solid curve is a fit with a BCS s-wave gap. For comparison a model assuming an additional small gap manifesting itself at low temperatures is shown as a dashed line.
$R_B$ for ultrathin FeSe layers on oxide substrates such as SrTiO$_3$, MgO and KTaO$_3$ and determine various length scales and critical thicknesses. Particularly, Hall measurements as a function of thickness allowed to determine the length scale of the charge distribution due to charge transfer from the substrate, $d_{CT}$, and the penetration length of the superconducting order parameter $\xi_{SN}^T$ in the layer above due to the proximity effect. For FeSe/STO $d_{CT} \approx 4$ nm and $\xi_{SN}^T \approx 3.5$ nm, implying that ultrathin FeSe may exhibit high-$T_c$ superconductivity on an effective length higher than that inferred by ARPES measurements of the electronic structure of $\geq 2$ ML FeSe. Even allowing for band bending effects increasing the thickness of the charge transfer layer in the specific electric dipole layer configuration of Ref. [20], it appears reasonable to consider that proximity effects cannot be ignored in $> 1$ ML thick FeSe layer. In this respect it is interesting to note that the value $n_s^{2D} \approx 1.4 \times 10^{14}$ cm$^{-2}$ obtained from the excess electron determination by ARPES is about a factor of four lower than the present determination of the superconducting carrier density $n_s^{2D} \approx 6 \pm 2 \times 10^{14}$ cm$^{-2}$ of our heterostructure containing 1+4 FeSe layers.

The temperature dependence of the superfluid density (Figure 8) may suggest an increase of this quantity at the lowest measured temperature, 5 K. Since this effect appears only in a single data point we can only speculate about its significance. It might point to the presence of a second (small) gap effectively opening below 10 K. We allow for band bending effects increasing the thickness of our heterostructure containing 1+4 FeSe layers.

Each component of equation 5 can be calculated within the local London approximation ($\lambda \gg \xi$) as

$$\frac{n_s^{2D}(T)}{n_s^{2D}(0)} = \frac{n_s^{2D}(T, \Delta_1(0))}{n_s^{2D}(0, \Delta_1(0))} + (1 - \lambda \frac{n_s^{2D}(T, \Delta_2(0))}{n_s^{2D}(0, \Delta_2(0))},$$

where $\lambda(0)$ is the value of the penetration depth at $T = 0$ K, $\Delta_i(0)$ is the value of the $i$-th ($i = 1$ or 2) superconducting gap at $T = 0$ K and $\omega$ is the weighting factor of the band with the largest gap.

The temperature dependence of the superfluid density (Figure 8) may yield for the main gap $\Delta(0) = 10.5(1.6)$ meV (in agreement with the single-gap fit) and the putative small gap $\Delta(0) = 1.3(6)$ meV with relative weight 0.23(4). Another possibility may be some proximity contribution of the additional 4 monolayers of FeSe modifying the gap structure. Further measurements are needed to elucidate this point, as well as the question about the possible presence of additional small gaps at much lower temperature and their nodal structure.

To conclude, by measuring ex situ the depth and temperature dependence of the local field distribution in a heterostructure containing a buried superconducting ultrathin FeSe layer, we detect the formation of a vortex state below $T_c \approx 60$ K and quantify the superfluid density of 1+4 ML FeSe. The temperature dependence can be well explained by a single BCS $s$-wave gap of 10.2(1.1) meV. The $\mu$SR spectra show that the vortex state and superconductivity are homogeneously formed across the entire interface over a sample with a sizeable amount of charges condensing below $T_c \approx 62$ K. This shows that superconductivity in the buried interface has stable character and that inhomogeneities or imperfections of the substrate or of the overlayers do not hamper the formation of a superconducting state nor sizably modify its properties. A very sensitive magnetic probe such as polarized muons do not see indication of static or dynamic magnetism. The simple structure of single-layer FeSe, its high $T_c$ with $s$-wave type of gap and rather clean BCS character make it an ideal system to develop a microscopic understanding of high-$T_c$ superconductivity.

Acknowledgments The $\mu$SR experiments were performed at the Swiss Muon Source, Paul Scherrer Institut, Villigen, Switzerland. We thank A. Suter for fruitful discussions.

* Pabitra.Biswas@stfc.ac.uk Current address: ISIS Pulsed Neutron and Muon Source, STFC Rutherford Appleton Laboratory, Harwell Campus, Didcot, Oxfordshire, OX11 0QX, UK

1 elvezio.morenzoni@psi.ch

J. G. Bednorz & K. A. Müller Possible high-$T_c$ superconductivity in Ba-La-Cu-O system, Z. Phys. D. 64, 189-193 (1986).

2 A. Schilling, M. Cantoni, J. D. Guo, & H. R. Ott, Superconductivity above 130 K in the Hg-Ba-Ca-Cu-O system. Nature 363, 56-58 (1993).

3 Y. Kamihara, T. Watanabe, M. Hirano, & H. Hosono, Iron-Based Layered Superconductor La[012]FeAs ($x = 0.05 - 0.12$) with $T_c = 26$ K. J. Am. Chem. Soc. 130, 3296 (2008).

4 F.C. Hsu, et al. Superconductivity in the PbO-type structure a-FeSe. Proc. Natl. Acad. Sci. U.S.A. 105, 14262-14264 (2008).
D.C. Johnston, The puzzle of high temperature superconductivity in layered iron pnictides and chalcogenides. *Adv. Phys.* **59**, 803-1061 (2010).

J. Paglione, R. L. & Greene, High-temperature superconductivity in iron-based materials. *Nat. Phys.* **6**, 645-658 (2010).

G. R. Stewart, Superconductivity in iron compounds. *Rev. Mod. Phys.* **83**, 1589-1652 (2011).

F. Wang, & D. H. Lee, The electron-pairing mechanism of iron-based superconductors. *Science* **332**, 200-204 (2011).

Q. Y. Wang, *et al.* Interface-Induced High-Temperature Superconductivity in Single-Unit-Cell FeSe Films on SrTiO$_3$. *Chin. Phys. Lett.* **29**, 037402 (2012).

J. J. Lee, *et al.* Interfacial mode coupling as the origin of the enhancement of $T_c$ in FeSe films on SrTiO$_3$. *Nature* **515**, 245 (2014).

D. F. Liu, *et al.* Electronic origin of high-temperature superconductivity in single-layer FeSe superconductor. *Nat. Commun.* **3**, 931 (2012).

S. L. He, *et al.* Phase diagram and electronic indication of high-temperature superconductivity at 65 K in single-layer FeSe films. *Nat. Mater.* **12**, 605-610 (2013).

S. Y. Tan, *et al.* Interface-induced superconductivity and strain-dependent spin density waves in FeSe/SrTiO$_3$ thin films. *Nat. Mater.* **12**, 634-640 (2013).

S. N. Rebec, *et al.* Coexistence of Replica Bands and Superconductivity in FeSe Monolayer Films, S. N. Rebec, T. Jia, C. Zhang, M. Hashimoto, D.-H. Lu, R. G. Moore, and Z.-X. Shen, Phys. Rev. Lett. **118**, 067002 (2017).

Z. Wang, C. Liu, Y. Liu and J. Wang High-temperature superconductivity in one-unit-cell FeSe films. *J. Phys.: Condens. Matter* **29**, 153001 (2017).

J. Shigai, *et al.* Electric-field-induced superconductivity in electrochemically etched ultrathin FeSe films on SrTiO$_3$ and MgO. *Nat. Phys.* **12**, 42 (2016). doi:10.1038/nphys3530

B. Lei *et al.*, Evolution of High-Temperature Superconductivity from a Low-$T_c$ Phase Tuned by Carrier Concentration in FeSe Thin Flakes. *Phys. Rev. Lett.* **116**, 077002 (2016).

W. Zhang, *et al.* Direct observation of high-temperature superconductivity in one-unit-cell FeSe films. *Chin. Phys. Lett.* **31**, 017401 (2014).

H. Ding *et al.*, High-Temperature Superconductivity in Single-Unit-Cell FeSe Films on Anatase TiO$_2$ (001) *Phys. Rev. Lett.* **117**, 067001 (2016).

C. H. P. Wen, *et al.* Anomalous correlation effects and unique phase diagram of electron-doped FeSe revealed by photoemission spectroscopy. *Nat. Commun.* **7**, 10840 (2016).

J. F. Ge, *et al.* Superconductivity above 100 K in single-layer FeSe films on doped SrTiO$_3$. *Nat. Mater.* **14**, 285-289 (2015).

Q. Fan, *et al.* Plain $s$-wave superconductivity in single-layer FeSe on SrTiO$_3$ probed by scanning tunneling microscopy. *Nat. Phys.* **11**, 946 (2015).

D. Huang, *et al.* Revealing the Empty-State Electronic Structure of Single-Unit-Cell FeSe/SrTiO$_3$. *Phys. Rev. Lett.* **115**, 017002 (2015).

W. Zhang *et al.* Interface charge doping effects on superconductivity of single-unit-cell FeSe films on SrTiO$_3$ substrates. *Phys. Rev. B* **89**, 060506(R) (2014).

Z. Zhang, *et al.* Onset of the Meissner effect at 65 K in FeSe thin film grown on Nb-doped SrTiO$_3$ substrate. *Sci. Bulletin* **60** (14), 1301-1304 (2015).

J. Shigai, T. Miyakawa, Y. Ito, T. Nojima, and A. Tsukazaki, Unified trend of superconducting transition temperature versus Hall coefficient for ultrathin FeSe films prepared on different oxide substrates, Phys. Rev. B **95**, 115101 (2017).

H. Saadaoui, Z. Salman, T. Prokscha, A. Suter, B.M. Wojek, E. Morenzoni, Zero-field Spin Depolarization of Low-Energy Muons in Ferromagnetic Nickel and Silver Metal *Physics Procedia* **30**, 164 (2012).

E. Morenzoni, *et al.* Nano-scale thin film investigations with slow polarized muons. *J. Phys.: Condens. Matter* **16**, S4583 (2004).

T. Prokscha, *et al.* The new mu E4 beam at PSI: A hybrid-type large acceptance channel for the generation of a high intensity surface-muon beam. *Nucl. Instrum. Methods Phys. Res., Sect. A* **595**, 317 (2008).

E. Morenzoni, *et al.* Low-energy $\mu$SR at PSI: present and future. *Physica B: Condensed Matter* **289**, 653 (2000).

Ch. Niedermayer, *et al.* Direct observation of a flux line lattice field distribution across an YBa$_2$Cu$_3$O$_{7-\delta}$ surface by low energy muons. *Phys. Rev. Lett.* **83**, 3932 (1999).

E. Morenzoni, *et al.* The Meissner effect in a strongly underdoped cuprate above its critical temperature. *Nat. Commun.* **2**, 272 (2011).

A. Di Bernardo *et al.* Intrinsic Paramagnetic Meissner Effect Due to $s$-Wave Odd-Frequency Superconductivity, *Phys. Rev. X* **5**, 041021 (2015).

F. Al Ma’Mari *et al.* Beating the Stoner criterion using molecular interfaces, *Nature* **524**, 69 (2015).

W. Eckstein, *Computer Simulations of Ion-Solid Interactions*, (Springer Verlag Berlin, Heidelberg and New York, 1991).

E. Morenzoni *et al.* Implantation studies of keV positive muons in thin metallic layers. *Nucl. Instr. and Methods Phys. Res., Sect. A* **192**, 254-266 (2002).

G. M. Luke, *et al.* Time-reversal symmetry breaking superconductivity in Sr$_2$RuO$_4$. *Nature* **394**, 558 (1998).

P. K. Biswas, *et al.* Evidence for superconductivity with broken time-reversal symmetry in locally noncentrosymmetric SrPtAs. *Phys. Rev. B* **87**, 180503(R) (2013).

R. A. Kubo, stochastic theory of spin relaxation. *Hyperfine Interact.* **8**, 731 (1981).

A. Suter , B. M. Wojek, Mursfit: A Free Platform-Independent Framework for $\mu$SR Data Analysis, *Physics Procedia* **30**, 69 (2012).

J. E. Sonier, J. H. Brewer, R. F. & Kiefl, $\mu$SR studies of the vortex state in type-II superconductors. *Rev. Mod. Phys.* **72**, 769 (2000).

J. R. Clem, Two-dimensional vortices in a stack of thin superconducting films: A model for high-temperature superconducting multilayers. *Phys. Rev. B* **43**, 7837 (1991).

E.H. Brandt, Ginsburg-Landau vortex lattice in superconductor films of finite thickness. *Phys. Rev. B* **71**, 014521 (2005).

G. Carneiro, E. H. & Brandt, Vortex lines in films: Fields and interactions. *Phys. Rev. B* **61**, 6370 (2000).

E.H. Brandt, Ginsburg-Landau vortex lattice in superconductor films of finite thickness. *Phys. Rev. B* **71**, 014521 (2005).

J. Pearl, Current distribution in superconducting films carrying quantized fluxoids. *Appl. Phys. Lett.* **5**, 65 (1964).

H. Padamsee, J. E. & Neighbor, C. A. & Shiffman, Quasi-particle phenomenology for thermodynamics of strong-
coupling superconductors. *J. Low Temp. Phys.* **12**, 387 (1973).

48 M. Tinkham, *Introduction to Superconductivity*. (McGraw-Hill, New York, 1975).

49 R. Prozorov, R. W. & Giannetta, Magnetic penetration depth in unconventional superconductors. *Supercond. Sci. Technol.* **19**, R41 (2006).

50 A. Carrington, F. & Manzano, Magnetic penetration depth of MgB$_2$. *Physica C* **385**, 205 (2003).