Electronic transport properties of superconducting FeSe thin films in a magnetic field

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Abstract. Superconducting $c$-axis oriented tetragonal FeSe films were grown on (001) MgO substrates. The transition temperature increased with film thickness and reached 10.5 K for a 2 µm film. The upper critical field, $B_{c2}$, was measured up to 14 T and then extrapolated to lower temperatures by an interpolation scheme based on Werthamer-Helfand-Hohenberg theory. The highest $B_{c2}(0)$ was obtained for the 2 µm film: 28.8 T and 24.7 T in parallel and normal field, respectively. The effect of thermally activated flux-flow on the film resistivity was analysed, and the activation energies ($U_0$) were extracted. At temperatures not far below $T_c$ and fields above 1 T we observed a clear logarithmic dependence of $U_0(B)$, which is an evidence for the 2D vortex matter. At lower fields and temperatures the vortex matter behaved 3D-like.

FeSe with a tetragonal lattice is one of the simplest iron-based superconductors with respect to its binary chemical composition and crystallographic structure consisting of quasi-2D Fe-Se layers ($a$-$b$ plane) separated by a $c$-axis lattice spacing of $\sim 5.5$ Å[1]. Recently we have shown that the evolution of 2D electronic transport in FeSe thin films gives rise to a superconductor-insulator quantum phase transition [2] as the film thickness was reduced from 2 µm to 1 nm. In the current work we focus in more detail on the properties of superconducting films of this series, especially in dc magnetic fields.

The FeSe films were prepared in situ by simultaneous magnetron sputtering of a sintered planar FeSe and a pure Se target onto 5×10 mm$^2$ (001) MgO substrates [3]. For all examined films the $\theta$-$2\theta$ x-ray diffraction (XRD) scans revealed (001) tetragonal FeSe structure with lattice constant $c$ between 5.526 Å and 5.548 Å [3]. Minor amount of (101) tetragonal FeSe phase was often present and the spurious (001) hexagonal FeSe phase was observed in some films. The film lattice exhibited a good off-plane alignment. The (001) rocking curve of a 0.5 µm thick film had a full width at half-maximum (FWHM) $\Delta\omega = 0.22^\circ$. $\Delta\omega$ increased with the film thickness up to $2.8^\circ$ for a 2 µm film. The in-plane lattice alignment examined by $\phi$-scan XRD revealed an in-plane orientational relationship with the (100) axis of tetragonal FeSe parallel to the (110) axis of MgO, see figure 1. In this case each three unit cells of (001) FeSe ($a = b = 3.76$ Å) are accommodated diagonally over two unit cells of (001) MgO ($a = b = 4.21$ Å), see figure 1b, resulting in a reasonably low 5% lattice mismatch.

The dc resistivity ($\rho$) of the FeSe films was measured using a Keithley 6220/2182 system as a function of temperature ($T$) in dc magnetic fields ($B$) up to 14 T directed either parallel or normal to the film surface. The measurements were performed by four-point technique over bridges patterned in the films. Figure 2 shows the whole set of resistivity data measured for a 2 µm FeSe film over a 200 µm
interested in the development of [4]. For a more detailed analysis we focused on the 2 µm film, see figure 3b. Most of all we were in-wide and 2 mm long bridge. At zero applied field this film exhibited low normal state resistivity, \( \rho_n(15K,0T) = 0.17 \, \text{m}\Omega\cdot\text{cm} \), which is comparable to bulk material [4]. In increasing field \( \rho_n \) grew at a rate of 0.3 \( \mu\Omega\cdot\text{cm/T} \) in parallel field and 1 \( \mu\Omega\cdot\text{cm/T} \) in normal field. Similar magnetoresistive behaviour has been reported for other Fe-based superconductors [5]. Above 15 K \( \rho_n \) was fairly linear with temperature and we used a linear extrapolation of \( \rho_n(T) \) for evaluating \( \rho(T)/\rho_n(T) \) ratios at \( T < T_c \). At no field applied the \( T_c \) onset was defined at 12.9 K (\( \rho(T)/\rho_n(T) = 0.9 \)), the \( T_c \) offset at 8.2 K (\( \rho(T)/\rho_n(T) = 0.1 \)) and the midpoint \( T_c \) at 10.5 K. These values are quite close to those of bulk material under ambient pressure, indicating (i) high film quality and (ii) that the film lattice was free of considerable mechanical stress. Increase of the magnetic field shifted the \( \rho(T) \) curves to lower temperatures but did not affect their shape: neither steps nor other weak-link induced features [6] appeared. Furthermore, the highest applied field of 14 T did not suppress the superconductivity to a remarkable extent. Thus we conclude the FeSe films to be free from distinct inhomogeneities.

Figure 3 shows the upper critical fields \( B_{c2} \) extracted from the data of figure 2 using the \( \rho(T)/\rho_n(T) = 0.9 \) criterion, as well as \( B_{c2}(T) \) data obtained for three other FeSe films of various thicknesses. In agreement with our former findings [2] thicker films exhibited higher \( T_c \). \( B_{c2} \) also increased with film thickness. \( B_{c2}(T) \) of the 2 µm film were very close to those reported for bulk FeSe at ambient pressure [4]. For a more detailed analysis we focused on the 2 µm film, see figure 3b. Most of all we were interested in the development of \( B_{c2}(T) \) at low temperatures. Usually, Werthamer, Helfand and Hohenberg (WHH) theory [7,8] is applied to predict \( B_{c2} \) at \( T = 0 \) K from the slope of \( B_{c2}(T) \) at \( T_c \) according to the WHH result \( B_{c2}(0) = 0.7 \, T_c \cdot |dB_{c2}(T)|/dT \). But this approach is only reliable for well-defined \( T_c \) and \( B_{c2}(T = T_c) \) data evaluation which is not really the case for our FeSe films: It leads to \( B_{c2}(0) = 61T \) for both parallel and normal field. However, since \( B_{c2}(T) \) data extracted well below \( T_c \) are much more reliable we derived an interpolation function \( B_{c2}(0K \leq T < T_c) \).

**Figure 1.** (a) The XRD \( \phi \)-scan on the (112) reflection of the FeSe film and (204) reflex of the MgO substrate and (b) the resulted in-plane orientation of the film on the substrate.

**Figure 2.** Resistivity of the 2 µm thick FeSe film in dc magnetic field applied parallel (a) and normal (b) to the film surface.
reported Maki parameters solved the WHH fixpoint equation numerically for the full range of the up to now experimentally magnetic field that the superconductor can tolerate below this orbital limit. In our calculations we first Wt on of the Pauli electron spin paramagnetism on the superconducting state. For t s, the weakening of the superconducting state reduces the maximum magnetic field that the superconductor can tolerate below this orbital limit. In our calculations we first solved the WHH fixpoint equation log[t] = F [t, b[t], α], where the functional F depends on t = T/Tc, b[t] = Borb(t)/Borb(T = 0 K) and the Maki parameter, α, quantifying the weakening influence of the Pauli electron spin paramagnetism on the superconducting state. For α = 0 this does not play any role and Borb(T) is given as the pure “orbital field limit” Borb(T) due to the supercurrents circulating around the vortex cores. For α > 0, the weakening of the superconducting state reduces the maximum magnetic field that the superconductor can tolerate below this orbital limit. In our calculations we first solved the WHH fixpoint equation numerically for the full range of the up to now experimentally reported Maki parameters 0 ≤ α ≤ 6 to obtain, for a given α, a set of solution points {b[t], t}. We then derived an analytic interpolation function ℎinterp[t; α] which approximates these sets of solution points within an accuracy of −1 % in the temperature range from 0 K to Tc and for 0 ≤ α ≤ 6. Our fit procedure allows to readily identify the reliable data range from which first α and then in the second step Borb(T = 0 K) and Borb(T = 0 K) are derived. More details of this evaluation scheme will be explained in a future publication. For Borb(T) of the 2 µm film we derived α|| = 1.7 for parallel and α⊥ = 0.4 for normal field. Such an anisotropy of the Maki parameter and the Pauli limiting field reflects the different paramagnetic influence on the Cooper pairs for the two different field orientations and is even more expressed in FeSe than in CeCoIn5 [9]: In the fields perpendicular to the Fe-Se layers a conventional pure orbital limitation prevails, whereas in B|| the Pauli limit is more dominant. The respective fits lead to the prediction B||(0) = 28.8 T and B⊥(0) = 24.7 T (see figure 3b). The conventional WHH estimate B||(0) = 61T actually has to be compared to our respective fit results B||(0) = 50 T and B⊥(0) = 25 T, demonstrating the error of the conventional B||(0) prediction method.

The ρ(T, B) data allow analysing the thermally activated flux-flow, which occurs above the irreversibility line and enforces ρ = ρ0 exp(-U0/kT), where U0 is the activation energy and k is the Boltzmann constant [10]. The slope of the Arrhenius plot, see figure 4, yields U0 for a given field B. However, for

**Figure 3.** (a) Borb(T) measured for the films of different thicknesses. (b) Data extrapolation to lower temperatures for the 2 µm film: the dashed and the dotted lines are fitted to the experimental data using our WHH functional fit for parallel and normal fields, respectively.

**Figure 4.** Data of figure 2b arranged in the Arrhenius plot. The dashed line at ρ/ρ0 = 0.1 illustrates the inapplicability of this criterion for the extraction of irreversibility fields of FeSe films, since the dissipative TAFF regime extends far below this line.
Figure 5. The activation energies extracted form the $\rho (T)$ at various $B_\perp$. (a) for the 2 $\mu$m thick FeSe film and (b) for the 0.5 $\mu$m thick FeSe film along with logarithmic and power-law fits.  

layered superconductors, e.g. oxygen-deficient YBa$_2$Cu$_3$O$_y$ [10], this slope may not remain constant: each curve is divided in two roughly linear parts with different slopes attributed either to a quasi-2D ‘pancake’ vortex matter at temperatures not far below $T_c$ with a logarithmic $U_0(B)$ dependence, or to a ‘conventional’ 3D vortex matter at lower temperatures with a power-law $U_0(B)$. For FeSe films these two ranges can be distinguished as well, see figure 4a. The extracted activation energies are assembled in figure 5a as $U_{0h}$ for the higher temperature range and $U_{0l}$ for the lower one. At higher temperatures the dissipation is likely due to 2D vortices: A logarithmic fit of $U_{0h}(B)$ appears more reasonable than any power-law fit. The classification of the lower temperature range is not unambiguous. For $B > 3$ T both fits describe $U_{0l}(B)$ well. Below $-2$ T the logarithmic fit may appear better, though the accuracy of $U_{0l}$ fades due to small resistivities. This unclear distinction between 2D an 3D behaviour can be explained by the lattice disorder accumulated over the 2 $\mu$m film growth and distorting the long-range order of FeSe planes (as noted above, the mosaic spread of the 2 $\mu$m film reached $\Delta \omega = 2.8^\circ$, compared to $\Delta \omega = 0.22^\circ$ of the 0.5 $\mu$m film). For comparison, $U_{0l}(B)$ for the 0.5 $\mu$m film is shown in figure 5b. The data of figure 5b also reveal a convincing logarithmic fit of $U_{0l}(B)$ extending to lower fields than in figure 5a, and the power-law fit of $U_{0l}(B)$ becomes distinctive. Apparently, high structural perfection is crucial for clear distinction between 2D and 3D vortex behaviour in FeSe films.  

Furthermore, the data of figure 4 imply the inapplicability of the $\rho (\rho_a = 0.1$ criterion for estimating the irreversibility fields for FeSe films since the TAFF region extends to much lower values of $\rho/\rho_a$.  

In summary, superconducting films of (001) tetragonal FeSe were grown on (001) MgO substrates with 5% lattice mismatch. Both $T_c$ and $B_{c2}$ of our films increased with the film thickness $d$ and reached at $d = 2$ $\mu$m the values reported for bulk under ambient pressure [4]. The concomitant reduction of the lattice alignment from $\Delta \omega = 0.22^\circ$ for $d = 0.5$ $\mu$m to $\Delta \omega = 2.8^\circ$ for $d = 2$ $\mu$m likely affected the electronic transport behaviour under application of magnetic field: The higher lattice misalignment smeared the distinction between 2D and 3D regimes of thermally activated flux-flow.

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