C-axis resistivity of superconducting FeSe single crystals: upper critical field and its angular behavior.

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Abstract

We report out-of-plane magnetotransport ρc(B, T) measurements for a high quality superconducting FeSe single crystals in magnetic fields up to 9 Tesla. Samples, grown from the flux under a permanent gradient of temperature with [001] crystallographic orientation were put in magnetic field parallel to ab-plane. The samples were rotated around c-axis, and its superconducting transitions R(H) were measured for each fixed angle in several temperatures. We show that Hc2 is anisotropic in these relatively small fields, with Hc2∥a/Hc2∥b being ~1.2 for T=8.3K.

Keywords: FeSe, upper critical field, superconductivity, single crystals

1 Introduction

Iron-based superconductors, discovered in 2008 (Y. Kamihara, 2008) remain in the forefront of scientific interest for last several years. These structures proved to be a pretty hard nut to crack, when it comes to the question of superconducting state symmetry and pairing mechanism. Even for the FeSe1-x system (and overall the “11” family), there is no general consensus on the nature of superconductivity despite the fact that “11” family has the simplest structure and is considered as a model system for iron pnictides and chalcogenides. Some papers give evidence to nodal superconductivity in FeSe1-x (C.-L. Song, 2011) with a help of surface-sensitive scanning tunneling spectroscopy. Bulk probes, such as specific heat (J.-Y. Lin, 2011) and thermal conductivity (J. K. Dong, 2009) point to the nodeless superconductivity. In paper (B. Zeng, 2010) a precise angle-
resolved heat capacity measurement in magnetic field show clear anisotropy in order parameter. Their experiment showed, that heat capacity oscillates with the angle of magnetic field orientation in the ab-plane. Since superconducting critical parameters such as $T_c$ and $H_{c2}$ are bound with structure of order parameter, we decided to research angular behavior of $H_{c2}$ in ab-plane.

In this paper we report experimental evidence for anisotropy in resistive upper critical field, when FeSe$_{1-x}$ sample is rotated in magnetic field around the c-axis.

2 Samples

The FeSe$_{1-x}$ single crystals were grown from the flux in a mixture of KCl and AlCl$_3$ under a permanent gradient of temperature in the SiO$_2$ ampoule. This method proved to be a reliable method for growing highly oriented FeSe$_{1-x}$ single crystals (D. Chareev, 2013). Similar method of crystal growth is sometimes referred to as vapor-growth method (A. E. Bohmer, 2013). The crystals have a mirror-like surface with the c-axis oriented perpendicular to the crystal plane, and have only a tetragonal $\beta$-FeSe phase present, without any peaks of hexagonal $\alpha$-FeSe phase. Figure 1 shows only (00L) reflexes. X-ray scanning was performed on both sides of a crystal, lattice parameter c from the bottom side $c_{bottom}=5.5226$ Å, is very close to value from the top side ($c_{top}=5.5225$ Å), proving that sample is highly uniform. Two other lattice parameters a and b differ from each other ($a=3.7713$ Å and $b=3.7736$ Å), meaning that tetragonal phase undergo a slight deformation, making it a little bit orthorhombic. This deformation and the difference between lattice parameters can be

![Figure 1: Temperature dependence of c-axis resistivity in zero magnetic field. A transition from tetragonal to orthorhombic phase is clearly visible at ~87K (shown with an arrow). Inset shows a sharp superconductive transition at $T_c=9.2K$.](image1.png)

![Figure 2: X-ray diffractogram of one of our samples, it shows only (00L) reflexes, from L=1 to L=4.](image2.png)
explained by different Se-vacancy concentration along axes a and b, because the real composition of the β-FeSe is FeSe$_{1-x}$, and the Se-vacancies in the crystal have a compression effect on lattice parameters. And Se-vacancy concentration along the a-axis is higher due to faster growth velocity as compared to the b-axis.

The transition from tetragonal phase to orthorhombic appears at 87K (Figure 2). Superconducting transition starts at 9.2K, and has ~0.6K width in zero field.

3 Experiment

In order to study angle resolved magnetotransport properties, one needs to be sure, that current in the sample is always perpendicular to the magnetic field. For this to be the case we carefully adjusted the sample orientation, so that ab-plane is parallel to magnetic field (with error less than 1 degree), and we used co-axial arc-shaped golden current contacts centered around point contacts for voltage (see Figure 4 inset).

For our experiments we used a Quantum Design PPMS Rotator insert. We measured resistive superconductive transitions in different fixed temperatures in several field orientations in ab-plane. For each temperature we changed orientation angle in one same direction so that to get rid of any mechanical backlash and we measured magnetoresistence $R_c(H)$ from -9Tesla up to +9Tesla, taking into account the possible addition of Hall resistance. Then the data was symmetrized and as a result we got a set of transition curves for all the angles and all temperatures. (Figure 3 shows one set of curves for T=8.3 K).

In order to determine $H_{c2}$ we used criterion of maximum differential of dR/dB from R(B) curves in Figure 3. It is a commonly known fact, that the criterion is essential for upper critical field determination, and different results (for example $H_{c2}$ temperature behavior) can be obtained when different criteria are used. Vedeneev et al. showed (S. I. Vedeneev, 2013) that in higher magnetic fields superconducting transitions become broad, making the choice of the criterion highly impactful on the resulting curves for $H_{c2}$, but in case of smaller fields (up to 10 Tesla for FeSe$_{1-x}$ all the different criteria data can be easily scaled to one figure.

![Figure 3: Field dependences of c-axis resistivity for various field orientations in ab-plane. T=8.3K](image)

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![Figure 4: Angular dependence of upper critical field for different temperatures. Inset shows the experimental scheme.](image)
reduced $H_{c2}(T)$ curve. That is why we mostly used only one criterion.

On Figure 4 we show the angular dependence of upper critical field for different temperatures. There are two main features, bound to crystallographic axes (shown by arrows a and b). This means, that even though the difference in crystallographic parameters of the orthorhombic FeSe phase is rather small, the $H_{c2}$ changes significantly in those two directions. Furthermore, a more subtle $H_{c2}(\theta)$ features can be seen between those two directions, but studying the nature of those features requires more angular resolution. Even more interesting is how the angular dependence evolves in the lower temperature region, but these experiments will need higher magnetic fields.

The origin of this upper critical field anisotropic behavior is unclear yet. Recent studies show that Fermi surface in FeSe significantly distorts and can lead to in-plane anisotropy in electronic properties in the low temperature region in both superconducting and normal states (M. D. Watson, 2015) (S-H. Baek, 2014). On the other hand the observed anisotropy of upper critical field can be explained by the anisotropic nature of order parameter in “11” systems (Abdel-Hafiez, 2013) (B. Zeng, 2010) (J.-Y. Lin, 2011). More detailed investigations with more angular resolution, complemented with magnetization and thermodynamic experiments are yet to be done in order to clarify the nature of ab-plane anisotropy.

4 Summary

Our results show, that there is significant anisotropy in upper critical field in the ab-plane, measured by $R_c(H)$ experiments with a sample rotating around c-axis in magnetic field. The higher value in $H_{c2}$ corresponds to b-axis, and a kink in $H_{c2}(\theta)$ dependence corresponds to the direction of a-axis. This $H_{c2}$ behavior can be explained by Fermi surface geometry, or by the superconducting order parameter anisotropy, or probably a combination of both. It is also worth noting, that the lack of consistency in the experimental results about anisotropy parameter $\Gamma=H_{c2||ab}/H_{c2||c}$ (Mahmoud Abdel-Hafiez, 2015) (S. I. Vedeneev, 2013) (V. A. Gasparov, 2011) (Alain Audouard, 2015) can be partially explained by the fact, that the value of $H_{c2||ab}$ is different with different field orientations in ab-plane.

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