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Vortex pinning by intrinsic correlated defects in $Fe_{1-y}Se$

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Abstract. We present a study on the transport and magnetic properties of superconducting $Fe_{1-y}Se$ single crystals. In the superconducting state, the in-plane electrical resistivity of the crystal is measured for fields up to 16 T and as a function of field direction, in order to understand how the vortex dynamics is affected by the presence of defects. A strong deviation from the slightly anisotropic crystal (electronic anisotropy constant $\gamma \sim 1.08$) is observed as a steep angular dependence, which is interpreted as a signature of the presence of correlated defects. The influence of the correlated defects on the critical current is studied through the angular dependence of the magnetization, and compared to numerical simulations.

1. Introduction

Recently, a new iron-based superconductor family was found, of which $FeSe$, the compound focused on here, is a member [1]. In all cases, the samples present a tetragonal layered structure and a slight anisotropy, $\gamma = m_c/m_{ab}$, with m_c and m_{ab} the effective electronic masses in the c axis and the ab plane, respectively [2].

It is common to find spurious phases in the crystals mixed with the superconducting tetragonal $FeSe$ since the latter only spans a very narrow region of the thermodynamic phase diagram [3]. The spurious phases could include metallic Fe , in excess of this element, or Fe_7Se_8 , in its defect. In this work we identify how the presence of Fe_7Se_8 affects the superconducting vortex dynamics in $Fe_{1-y}Se$ single crystals.

2. Crystal characterization

$Fe_{1-y}Se$ single crystals were grown from 1/4 KCl :3/4 $NaCl$ flux in evacuated double quartz ampoules. Structural characterization was made by X Ray diffraction (XRD). Chemical composition was quantified by Energy Dispersive Spectroscopy (EDS). The crystals have a platelet shape and present Fe deficiency. XRD shows the presence of two phases, the superconducting tetragonal $FeSe$, and the ferrimagnetic hexagonal Fe_7Se_8 . Tetragonal $FeSe$ shows two preferential orientations of the c axis, one parallel and the other at 57° from the platelet normal. Hexagonal Fe_7Se_8 grows only with the c axis parallel to the normal [4] and presents an ordered array of Fe vacancies [5].



3. Results and discussion

Figure 1 shows in plane electrical resistivity, ρ , measurements for an $Fe_{0.92}Se$ crystal with magnetic field, H , perpendicular to the crystal plane. The onset of the transition temperature for $H = 0$ is $T_c = (11.9 \pm 0.1)$ K. The transition width is broadened by about 35% when H varies from 0 to 16 T. This is a signature for the presence of vortex liquid.

In order to study the influence of correlated defects on the dynamics of the vortex lattice we measured the angular dependence of ρ in terms of the orientation of H . The angle of H is measured with respect to the normal to the platelet, see θ in Fig. 2a). We used a constant Lorentz force configuration, i.e. the external field is always at a right angle to the applied current. This avoids spurious angular dependencies of the pinning force related to the non-perpendicularity between current density and magnetic field. Figures 2b) and 2c) show the

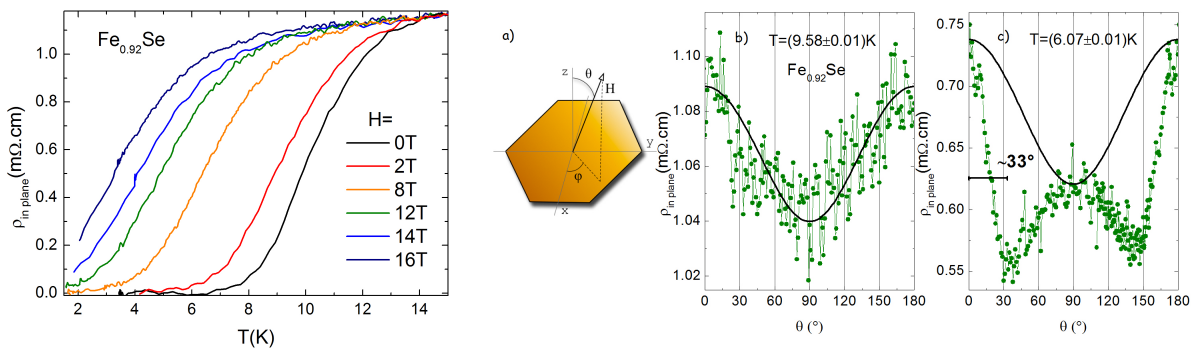


Figure 2. a) Definition of the rotation angle, θ (here $\varphi = 0$ and transport current is along y). b) and c) Angular dependence of the in plane resistivity for an applied field of $H = 12$ T, and temperatures $T = (9.58 \pm 0.01)$ K and $T = (6.07 \pm 0.01)$ K respectively. The solid lines represent the expected behavior for an anisotropic material with point defects for $\gamma = (1.08 \pm 0.05)$.

Figure 1. Temperature dependence of the in plane resistivity for a crystal of $Fe_{0.92}Se$ with magnetic field perpendicular to the crystal plane.

results for a $Fe_{0.92}Se$ crystal at $H = 12$ T and for $T = (9.58 \pm 0.01)$ K and $T = (6.07 \pm 0.01)$ K. The continuous line shows the expected behavior for a slightly anisotropic material with point defects. This dependence was evaluated in base to a thermally activated ρ [6] and to the scaling for the angular dependence proposed by Blatter and co-workers [7]. The final equation is:

$$\rho(T, H, \theta) = \frac{\rho_0}{T} \exp \left[-U(T, H, \theta) T^{-1} \right] \quad \text{with} \quad U(T, H, \theta) = U_o(H\varepsilon(\theta))^{\frac{1}{2}-q} \left(1 - \frac{T}{T_c} \right)^q \quad (1)$$

where $\varepsilon(\theta) = \sqrt{\cos^2\theta + \gamma^{-2}\sin^2\theta}$, U is the activation energy for the movement of the vortices, ρ_0 is a constant and q is a parameter that takes values between 0.5 and 2. In figure 2, we use $H = 12$ T, $T_c = (11.9 \pm 0.1)$ K and the corresponding temperature. The parameters $\rho_0 = (13 \pm 2)$ m Ω .cm.K, $q = (1.1 \pm 0.2)$, $U(9.58 \text{ K}, 12 \text{ T}, 0^\circ) = (51 \pm 4)$ K and $U(6.07 \text{ K}, 12 \text{ T}, 0^\circ) = (141 \pm 6)$ K were obtained adjusting $\rho(T, H, \theta = 0^\circ)$ in Fig. 1. Finally, for both temperatures, we get $\gamma = (1.08 \pm 0.05)$ fitting Eq. 1 to the data in Fig. 2, and using the former values for the other parameters.

Notice that at temperatures near T_c the angular dependence is very well described by the equation above. However, for lower temperature, the resistivity presents strong deviations from this behavior. This is observed in all samples, and can be understood in terms of vortex dynamics. At low temperature, the extra minimum at $\pm 33^\circ$ (Fig. 2c)) could signal vortex pinning by correlated defects. The absence of this minimum near the critical temperature (Fig.

2b)) is consistent with the idea that vortices move in the flux flow regime and the pinning is less effective.

It is also expected for the correlated defect pinning to be observed in the critical current which should in turn, be observable with magnetization measurements. In order to assess these effects, we measured the longitudinal ($M_L \parallel H$) and transversal ($M_T \perp H$) magnetization of the crystals as a function of the rotation angle θ around an horizontal axis, while keeping the modulus of H constant, for $H = 0.15$ T at 2 K, well below T_c , and at 20 K, around 10 K above T_c .

In these crystals, there are two contributions to the magnetization, one coming from the superconducting $FeSe$, and the other coming from the ferrimagnetic Fe_7Se_8 . In order to separate both signals, we assume that they are independent, and that the magnetization of Fe_7Se_8 is approximately constant in our measurements, given that the Neel temperature is much higher ($T_N \sim 455$ K) [5]. Consequently, we obtain the superconducting $FeSe$ contribution to the magnetization by simply subtracting the 20 K curve from the 2 K one.

Figure 3 depicts the results in a plot of $M_i = \pm\sqrt{M_L^2 + M_T^2}$ as a function of the field component normal to the platelet, $H_z = H\cos\theta$, showing the superconducting magnetization loop. The curve has a rich structure due to the contribution of anisotropy, correlated defect pinning and crystal shape. In particular, at the angles where extra minima are observed in ρ , i.e. 33° , 147° , 213° and 327° , the magnetization loop also presents distinctive structure. Theoretically, this structure can be understood in terms of a critical current density with anisotropic behavior. Figure 4 summarizes the main facts in a number of simulations that were performed by a critical state modelling [8]. A θ -rotation experiment for an isotropic platelet-like superconductor would produce a *bumped* cycle as compared to the conventional *Bean-like* behavior for a standard H_z experiment. An extra magnetic moment occurs at $\theta \approx 90^\circ$. This is a purely geometric effect related to the *force free* configuration (due to the zones with induced current parallel to the magnetic field). On the other hand, when an anisotropic

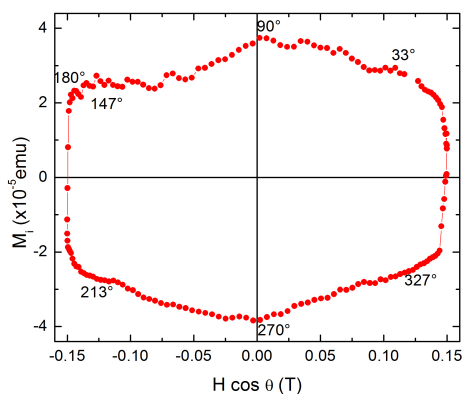


Figure 3. Superconducting contribution to the magnetization, $M_i = \pm\sqrt{M_L^2 + M_T^2}$, as a function of $H\cos\theta$ for $H = 0.15$ T and $T = 2$ K for a crystal of $Fe_{1-y}Se$.

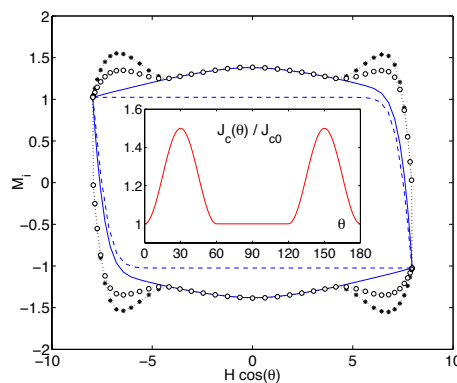


Figure 4. Same as Figure 3, but obtained from critical state simulations. The dashed line corresponds to a conventional Bean model cycle, the continuous line is for the platelet rotating in a magnetic field assuming isotropic critical current. Symbols are obtained by using anisotropic J_c as shown in the inset. A maximum $J_c(\theta)$ of 1.5 and 2.0 was used respectively. In all cases, units for $H\cos\theta$ are $J_c/4\pi$ and units for M_i , $J_c(a/d)$ with a the radius and d the thickness of the platelet.

critical current $J_c(\theta)$ enhanced at $\pm 30^\circ$ is assumed, a superimposed structure appears, with new features at $30^\circ, 150^\circ, 210^\circ, 330^\circ$ as observed in experiments. The more anisotropic $J_c(\theta)$, the more pronounced the effects.

In relation to the crystal microstructure, the correlated defects observed in transport and magnetic measurements may be linked to the phase mixture and the growth directions of the tetragonal phase. Figure 5 presents the crystal structure scheme of our samples, panel a) shows the possible directions of the c axis of the tetragonal phase compatible with our XRD measurements. The direction of the ab plane at $\sim 34^\circ$ from the normal of the crystal is coincident with a dense plane of vacancies in Fe_7Se_8 as can be inferred from the structural study of ref. [5] (see Fig. 5b)). This plane is, possibly, the phase boundary between the tetragonal and hexagonal phases and it could behave as a correlated defect for the vortex lattice.

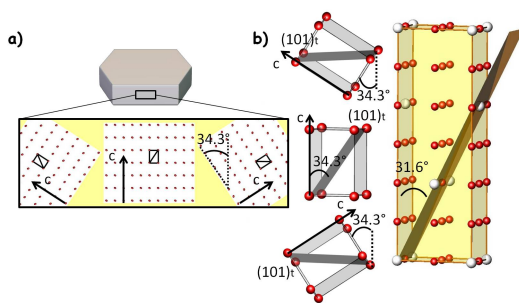


Figure 5. Crystal structure scheme of our samples. a) Possible directions of the c axis of the tetragonal phase. b) Unit cell of the tetragonal phase (*Left*) with the different orientations of the c axis and the unit cell of the hexagonal phase (*Right*). We only show the Fe atoms in red and the vacancies in white.

4. Conclusion

Electrical resistivity and magnetization results in $Fe_{1-y}Se$ crystals show clear evidence for the existence of superconducting pinning by correlated defects. At the same time, the structural characterization of the samples shows the existence of different phases and orientations. One of the crystalline orientations of the $FeSe$ phase coincides with the direction in which the correlated defects increase the pinning, evidencing that the origin of the defects could be related to phase matching in the samples.

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