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# Magnetic field - dependent pinning potential in LiFeAs superconductor from Campbell penetration depth

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A theoretical critical current density,  $j_c(T, H)$ , as opposite to commonly measured relaxed persistent (Bean) current,  $j_B$ , was extracted from the Campbell penetration depth,  $\lambda_C(T, H)$  measured in single crystals of LiFeAs. The effective pinning potential is slightly non-parabolic, which follows from the magnetic field - dependent Labusch parameter  $\alpha$ . At the equilibrium (upon field - cooling),  $\alpha(H)$  is non-monotonic, but it is monotonic at a finite gradient of the vortex density. Combined with the observation of a “fishtail” magnetization in standard DC measurements, this result implies that “fishtail” appears as a result of magnetic relaxation. The functional form of  $M(H)$  curves is determined by the non-monotonic pinning potential, implying the importance of vortex collective effects. The values of  $j_c(2\text{ K}) \simeq 1.22 \times 10^6$  A/cm<sup>2</sup> provide an upper theoretical estimate of the current carrying capability of LiFeAs. Overall, vortex behavior of almost isotropic, fully-gapped LiFeAs is very similar to highly anisotropic d-wave cuprate superconductors, the similarity that requires further studies in order to understand unconventional superconductivity in cuprates and pnictides.

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The determination of the critical current density  $j_c$ , is one of the fundamental problems in the vortex physics of type-II superconductors. Not only it is important for the assessment of the current-carrying capabilities relevant for practical applications, but knowing theoretical  $j_c$  is needed to understand microscopic mechanisms of vortex pinning. What is often called “critical current” is routinely determined from conventional DC magnetization measurements, alas this quantity is a convolution of theoretical  $j_c$  and magnetic relaxation during the characteristic time,  $\Delta t$ , of the experiment. For example, in case of ubiquitous *Quantum Design* MPMS (SQUID) magnetometry,  $\Delta t \geq 10$  sec. We will call measured supercurrent  $j_B$  to distinguish it from the theoretical  $j_c$  that is achieved when the vortices are de-pinned by the Lorentz force. By definition,  $j_c$  is reached when the energy barrier for vortex motion vanishes,  $U(j_c) = 0$ , whereas the measured current density  $j_B$  is determined by  $U(j_B) = k_B T \ln(1 + \Delta t/t_0)$ , where  $t_0 \lesssim 1$   $\mu$ sec is the characteristic time scale that depends on both sample geometry and details of pinning<sup>1-5</sup>. This also results in a quite different temperature dependence of  $j_B(T)$  compared to  $j_c(T)$ . Another approach to measure critical current density is to use AC susceptibility. Conventional time-domain susceptometers operate at frequencies  $f \lesssim 10$  kHz (hence  $\Delta t \gtrsim 0.1$  msec) and have large driving amplitudes,  $H_{ac} \gtrsim 0.1$  Oe. Such perturbation displaces vortices from the potential wells and one can use harmonics analysis to determine frequency - dependent current density,  $j_B(T, B, f)$ . This technique has been applied in both global<sup>6</sup> and local<sup>7,8</sup> forms.

In Fe-based superconductors flux creep is substantial

at all temperatures, thus measured  $j_B$  is expected to be lower than  $j_c$ . Indeed, reports produce only moderate current densities,  $j_B \lesssim 10^6$  A/cm<sup>2</sup>, - unusual for low-anisotropy high- $T_c$  materials<sup>9-15</sup>.

To access the information about pinning potential itself, one needs to measure the linear response when vortices are not driven out of the pinning potential wells. One way to do this is to measure so-called Campbell penetration depth which determines how far a small AC magnetic field penetrates the superconductor in the presence of vortices (induced by static external magnetic field) in the limit of  $H_{ac} \rightarrow 0$ , when vortex response is purely elastic and linear<sup>16-18</sup>. For a pinning potential,  $V(r)$ , the vortex displacement from the equilibrium position due to small  $H_{ac}$  is found from  $dV/dr = f_L$ , where the Lorentz force,  $f_L = j \times \phi_0/c$ . Maximum force determines the theoretical critical current density,  $j_c = car_p/B$ , attained at the range of the pinning potential  $r_p$ . If vortex distribution is inhomogeneous, static (Bean) current<sup>19</sup>,  $j_B$ , is superimposed with the excitation AC current and the response is determined by the effective Labusch constant  $\alpha(j_B) \equiv d^2V/dr^2|_{r=r_0}$ . Clearly  $\alpha(j_B)$  is constant only for a parabolic  $V(r)$ . The Campbell penetration depth is given by  $\lambda_C^2 = \phi_0 B / (4\pi\alpha(j_B))$ <sup>16-18,20</sup>.

Consider a typical experiment, which we use in the following. Sample is cooled in zero magnetic field and then static magnetic field is applied. This creates a gradient of vortex density supported by the persistent Bean current density  $j_B$ <sup>19</sup>. Small-amplitude  $H_{ac}$  causes vortex vibrations within pinning potential well, a condition for Campbell penetration depth measurements<sup>16-18,20</sup>. After the sample is warmed above  $T_c$  it is cooled again keep-

ing external static field constant (field-cooling) whence  $j_B = 0$ . We therefore may expect some hysteresis with  $\lambda_{C,ZFC} > \lambda_{C,FC}$  if  $V(r)$  is non-parabolic. Therefore, by measuring zero field - cooled (zfc) field-cooled (fc)  $\lambda_C$  at different magnetic fields and temperatures we can estimate theoretical  $j_c(H, T)$  and access the information regarding shape of the pinning potential. For more details the reader is referred to earlier studies of high  $-T_c$  cuprates<sup>20</sup>.

One of the most interesting and commonly observed features of unconventional superconductors is so-called second magnetization peak (also known as “fishtail”)<sup>3</sup>. It has now been observed in most Fe-based superconductors when magnetic field is aligned parallel to the crystallographic  $c$ -axis<sup>9,10,12–15,21</sup>. The origin of “fishtail” can be static, i.e., when theoretical  $j_c(H)$  is a non-monotonic function of field,  $H$ , or it can be dynamic caused by field-dependent magnetic relaxation<sup>5,22</sup>. Experimental determination of the origin of the “fishtail” in each material is, thus, very important as it allows to shed light on the nature of the flux pinning, hence defect structure “seen” by the Abrikosov vortices. In Fe-based superconductors, the interest is further fueled by multiple reports that defects, even non-magnetic, are pair-breaking due to, presumably, unconventional  $s_{\pm}$  symmetry of the order parameter<sup>23,24</sup>. Additionally, it seems that low-field behavior of most pnictides is governed by the so-called strong pinning, which results in a sharp peak in magnetization at  $H \rightarrow 0$ <sup>21</sup>. Therefore, to conduct a clean, baseline experiment, one ideally needs Fe-based superconductor with reduced scattering. These materials are rare, but do exist in form of only few stoichiometric compounds, LiFeAs being one of them. Due to high sensitivity to air and moisture, there are only few reports on the vortex properties in LiFeAs crystals. “fishtail” effect and relatively high  $j_B$  (5 K)  $\approx 1 \times 10^5$  A/cm<sup>2</sup> were found in Ref.[15], whereas much lower  $j_B$  (5 K)  $\approx 1 \times 10^3$  A/cm<sup>2</sup> was reported in Ref.[25]. Such spread may be related to clean - limit superconductivity in this compound when even small variation of impurity concentration causes significant change in the persistent current density and magnetic relaxation.

In this paper we report measurements of Campbell penetration depth in single crystals of LiFeAs. We show that the “fishtail” is revealed as a result of magnetic relaxation. Its shape is derived from the transformation of the pinning potential itself with the applied field. Namely, Labusch constant (and “theoretical” critical current,  $j_c(H)$ ) is a monotonic function of field when Bean current (macroscopic vortex density gradient) is present, but it becomes a non-monotonic function of field at a homogeneous distribution of vortices. The values of  $j_c(2\text{ K}) \approx 1.22 \times 10^6$  A/cm<sup>2</sup> provide upper theoretical estimate of the current carrying capability of this material and show the significance of magnetic relaxation. We also find evidence for the strong pinning regime at the low fields. With the increase of the magnetic field vortex pinning and creep change to a collective regime

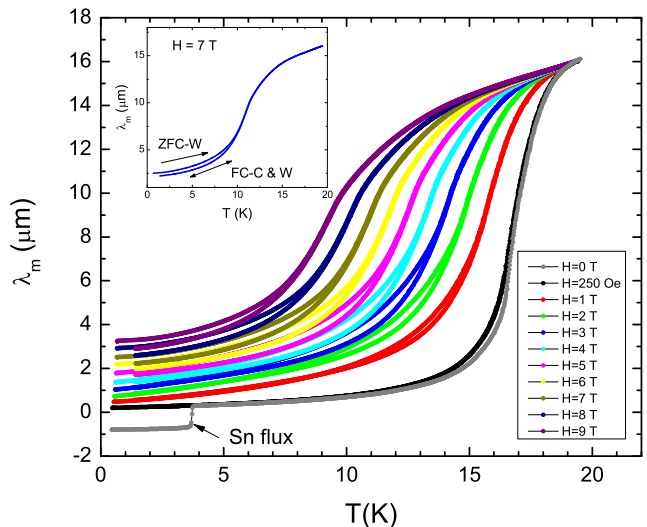


FIG. 1. (Color online) Magnetic penetration depth measured in a ZFC-FC process at different fields.  $H = 0$  curve shows a step due to leftovers of Sn flux. It was quenched by applying a  $H = 250$  Oe field. Inset shows an example of the small hysteresis of  $\lambda_m(T)$  at  $H = 7$  T.

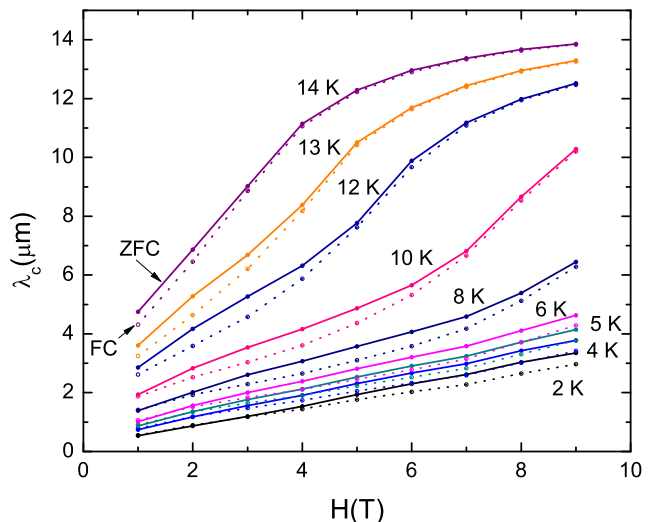


FIG. 2. (Color online) Campbell penetration depth as function of magnetic field at different temperatures extracted from the data of Fig. 1. Solid lines - ZFC and dashed lines are FC data.

and, finally, cross over to another vortex state, perhaps dominated by plastic deformations. Despite being quite different from high- $T_c$  cuprates in terms of pairing and gap structure, it seems that vortex behavior of Fe-based superconductors is remarkably similar to high- $T_c$  materials.

Single crystals of LiFeAs were grown out of Sn flux as described in detail elsewhere<sup>26</sup> and were transported for measurements in sealed ampoules. Immediately after opening,  $(0.5-1) \times (0.5-1) \times (0.1-0.3)$  mm<sup>3</sup> samples were

placed into the cryostat for the measurements. Additionally, samples were extensively characterized by transport and magnetization measurements<sup>26</sup>. Zero-field transition temperature of our samples was about,  $T_c \approx 18$  K. The magnetic penetration depth was measured with the tunnel - diode resonator technique (for review, see<sup>27</sup>). The sample was inserted into a 2 mm diameter copper coil that produced an rf excitation field (at  $f \approx 14$  MHz) of  $H_{ac} \sim 20$  mOe. An external DC magnetic field (0 – 9 T) was applied parallel to the AC field, both parallel to the  $c$ - axis,  $H_{ac} \parallel H \parallel c$ -axis. The shift of the resonant frequency (in cgs units) is given by  $\Delta f(T) = -G4\pi\chi(T)$ , where  $\chi(T)$  is the differential magnetic susceptibility,  $G = f_0V_s/2V_c(1 - N)$  is a calibration constant,  $N$  is the demagnetization factor,  $V_s$  is the sample volume and  $V_c$  is the coil volume. The constant  $G$  was determined from the full frequency change by physically pulling the sample out of the coil. With the characteristic sample size,  $R$ ,  $4\pi\chi = (\lambda/R) \tanh(R/\lambda) - 1$ , from which  $\Delta\lambda$  can be obtained<sup>27,28</sup>. The measured penetration depth consists of two terms, London penetration depth and Campbell penetration depth,  $\lambda_m^2 = \lambda_L^2 + \lambda_C^2$  [17]. Note that measured penetration depth does not diverge at  $T_c$ , because it reaches the limiting value determined either by the size of the sample or the normal skin depth, whichever is smaller. Due to pronounced temperature dependence above  $T_c$ , it seems that in our case it is skin-depth limited. We determined  $\lambda_L(T)$  from the measurements at  $H = 0$  and used a well-established value of  $\lambda_L(0) = 200$  nm [29].

Figure 1 shows magnetic penetration depth measured upon warming, after sample was cooled in zero field and target field was applied at low temperature (ZFC-W) compared to the measurements upon cooling when target field was fixed above  $T_c$  and kept constant (FC-C). A step at low temperatures on a  $H = 0$  curve is due to residual Sn flux. It was quenched by applying a moderate  $H = 250$  Oe field, which does not affect our analysis of the much higher fields. Inset in Figure 1 shows an example of the small magnetic hysteresis measured at  $H = 7$  T (notice that once ZFC-W process was complete, subsequent warming-cooling measurements (FC-C and FC-W) resulted in the same curve indicating homogeneous vortex distribution). The hysteresis between ZFC-W and FC-C-W is much smaller than, for example, observed in BSCCO crystals<sup>20</sup>, which is most likely due to much more 3D electronic nature of LiFeAs and it means that we can safely use the parabolic approximation of the pinning potential. From the measured penetration depth in zero field,  $\lambda_L(T)$ , and the one measured in applied magnetic field,  $\lambda_m(T, H)$ , we determine the Campbell penetration depth via,  $\lambda_C = \sqrt{\lambda_m^2 - \lambda_L^2}$  as shown in Fig.2.

From the Campbell penetration depth we determine the theoretical critical current density as,  $\frac{4\pi}{c}j_c = r_p\phi_0/\lambda_C^2$  were we assumed the radius of the pinning potential be a coherence length,  $r_p \simeq \xi \simeq 4.4$  nm. This estimate for  $\xi$  comes from the measurements of the upper critical field  $H_{c2}(0) \simeq 17$  T [30], but  $\xi \simeq 7$  nm

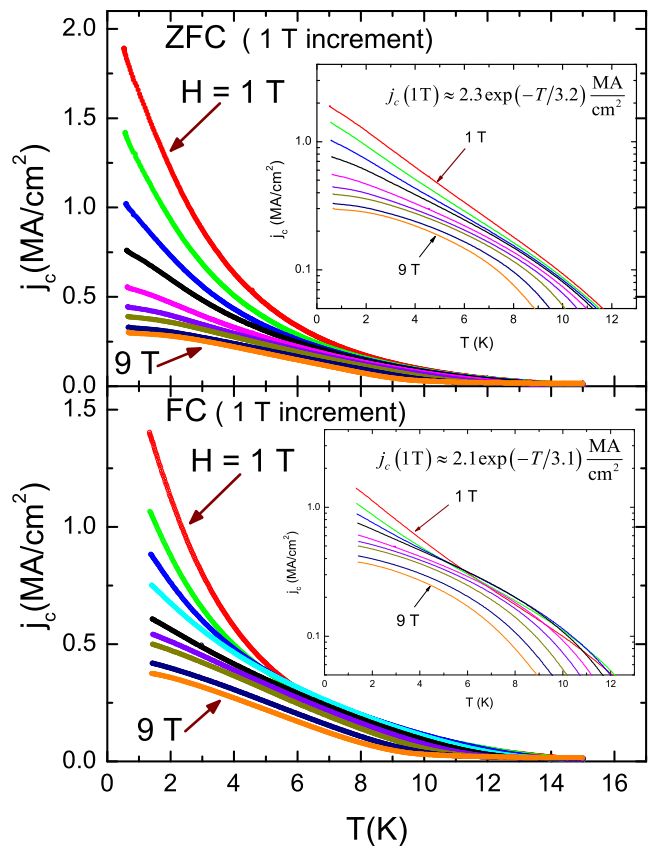


FIG. 3. (Color online) theoretical critical current density,  $j_c$ , determined from the ZFC (top frame) and FC (bottom frame) experiments at indicated values of the applied external magnetic fields. Insets show semi-log plots indicating exponential dependence of  $j_c$  at lower fields and a crossover to a different pinning regime at the higher fields.

has been reported from neutron scattering form factor<sup>31</sup>. Figure 3 shows  $j_c$  as a function of temperature at different magnetic fields determined after ZFC-W process (top frame) and FC-C process (bottom frame). In both cases, the curves are monotonic in temperature and show substantial temperature dependence similar to high  $-T_c$  cuprates, re-enforcing the earlier statement that vortex properties of Fe-based superconductors are remarkably similar to the cuprates, despite the difference in dimensionality of the electronic structure<sup>32</sup>.

To understand the functional dependence, we plot determined  $j_c(T)$  on a semi-logarithmic plot as shown in the insets in Fig. 3. At relatively low fields, the behavior is very similar to the earlier reports of strong pinning<sup>21</sup> and can be well approximated by the exponential temperature dependence,  $j_c(1\text{ T}) \simeq 2.1 \exp(-T/3.1)$  MA/cm<sup>2</sup> for FC-C process and  $j_c(1\text{ T}) \simeq 2.3 \exp(-T/3.2)$  MA/cm<sup>2</sup> for ZFC-W measurements. This very similar behavior imply that strong pins result in a more-or less parabolic  $V(r)$  and are practically independent of the bias Bean current,  $j_B$ . However, at the higher fields, the critical current becomes less temperature dependent,

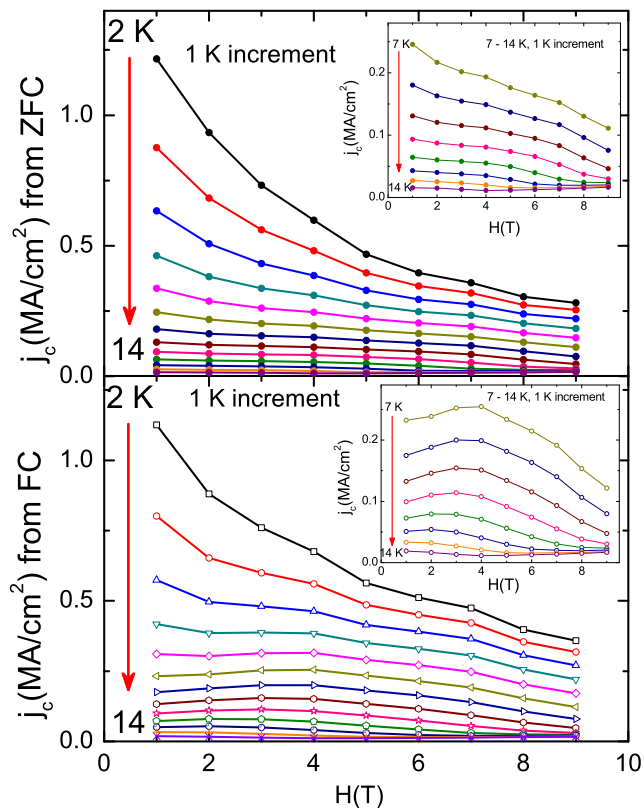


FIG. 4. (Color online) Critical current density determined from ZFC Campbell penetration depth (top frame) and from the FC Campbell penetration depth (bottom frame) showing the absence of the “fishtail” magnetization in the former and its presence in the latter.

probably due to saturation of strong pins and a crossover first to the collective pinning regime and eventually to the disordered lattice dominated by plastic deformations.

Finally, Fig. 4 shows “theoretical” critical current density,  $j_c$ , determined from ZFC Campbell penetration depth (top frame) and from the FC Campbell penetration depth (bottom frame) as a function of magnetic field at different temperatures. While ZFC curves are monotonic, a clear “fishtail” signature is observed in the equilibrium FC-C-W measurements at higher temperatures. The inset in Fig.4 emphasizes this result.

Our results can be interpreted in the following way. Estimated theoretical critical current density,  $j_c(2\text{ K}) \approx 1.22 \times 10^6 \text{ A/cm}^2$ , shows that conventional measurements probe under - critical currents, most likely due to significant magnetic relaxation. However, the most striking result is that  $j_c$ , obtained in a non-equilibrium ZFC process, is monotonic with the applied magnetic field at all temperatures, whereas equilibrium  $j_c$ , obtained in the FC process when magnetic flux distribution inside the sample is uniform, shows a clear signature of the “fishtail” (second peak) magnetization. (Note that FC  $j_c$  is only a convenient parameter characterizing the pinning potential and does not represent the current density that

can be measured). To relate our measurements of the local curvature of the pinning potential to the static problem of the maximum restoring force, we recall that for the determination of the Campbell length, the potential  $V(r)$  is Taylor - expanded around the bias point,  $r_0$ , so that  $V(r) \sim \alpha(r_0)(r - r_0)^2/2$ . The restoring force,  $dV/dr$  reaches maximum at the range of the pinning potential  $r_p$ , which determines the true critical current density that would actually be measured without magnetic relaxation. While  $r_0$  is somewhat less than  $r_p$ , the field dependence of  $\alpha(r_0)$  is monotonic and we therefore expect the true critical current density be monotonic with the magnetic field. Since conventional (relaxed) DC measurements show “fishtail” effect<sup>15</sup>, we conclude that this effect is of dynamic origin. The functional form of the  $M(H)$  curve is governed by the non-monotonic field - dependent pinning potential implying the importance of vortex collective effects. More specifically, with the decrease of a magnetic field, pinning potential  $V(r)$  at  $r = 0$  becomes more shallow, implying that the effective barrier for magnetic relaxation decreases. This is compatible with the collective creep model where “fishtail” develops as a result of magnetic relaxation<sup>3,5</sup>. It is possible that the origin of a “fishtail” in LiFeAs is similar to high - temperature cuprates. The question is how to reconcile a very different (almost isotropic) electronic properties of Fe-based superconductors and quite similar to highly anisotropic cuprates vortex behavior.

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