Magnetic-field-dependent pinning potential in LiFeAs superconductor from its 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 nonparabolic, which follows from the magnetic-field-dependent Labusch parameter \( \alpha \). At the equilibrium (upon field cooling), \( \alpha(H) \) is nonmonotonic, 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 the fishtail appears as a result of magnetic relaxation. The functional form of \( M(H) \) curves is determined by the nonmonotonic pinning potential, implying the importance of vortex collective effects. The values of \( j_c(2 \, \text{K}) \simeq 1.22 \times 10^6 \, \text{A/cm}^2 \) 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 is it important for the assessment of the current-carrying capabilities relevant for practical applications, but also 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 the case of the ubiquitous Quantum Design magnetic property measurement system, \( \Delta t \gtrsim 10 \, \text{s} \). We will call the measured supercurrent \( j_B \) to distinguish it from the theoretical \( j_c \) that is achieved when the vortices are depinned 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 \( U(j_B) = k_B T \ln(1 + \Delta t/\tau_0) \), where \( \tau_0 \lesssim 1 \, \mu\text{s} \) is the characteristic time scale that depends on both sample geometry and details of pinning.1–5 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 \( f \lesssim 10 \, \text{kHz} \) (hence, \( \Delta t \gtrsim 0.1 \, \text{ms} \) and have large driving amplitudes \( H_{ac} \gtrsim 0.1 \, \text{Oe} \). 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 global6 and local7,8 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, \( j_B \lesssim 10^6 \, \text{A/cm}^2 \), unusual for low-anisotropy high-\( T_c \) materials.9–15

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 a static external magnetic field) in the limit of \( H_{ac} \to 0 \), when vortex response is purely elastic and linear.16–18 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 \( j_c = carp/B \), attained at the range of the pinning potential \( f_p \). If vortex distribution is inhomogeneous, static (Bean) current19 \( j_B \) 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 \( \lambda_C^2 = \phi_0 B/[4\pi \alpha(j_B)] \).16–18,20

Consider a typical experiment, which we use in the following. A sample is cooled in a zero-magnetic field, and then, a static-magnetic field is applied. This creates a gradient of vortex density supported by the persistent Bean current density \( j_B \). Small-amplitude \( H_{ac} \) causes vortex vibrations within the pinning potential well, a condition for Campbell penetration depth measurements.16–18,20 After the sample is warmed above \( T_c \), it is cooled again by keeping the external static field constant (field cooling), whence, \( j_B = 0 \). Therefore, we may expect some hysteresis with \( \lambda_C, ZFC > \lambda_C, FC \) if \( V(r) \) is nonparabolic. Therefore, by measuring a zero-field-cooled (ZFC) field cooled (FC) \( \lambda_C \) at different magnetic fields and temperatures, we can estimate theoretical \( j_c(H, T) \) and can access the information regarding the shape of the pinning potential. For more details, the reader is referred to earlier studies of high-\( T_c \) cuprates.20
One of the most interesting and commonly observed features of unconventional superconductors is the so-called second magnetization peak (also known as fishtail). It has now been observed in most Fe-based superconductors when a magnetic field is aligned parallel to the crystallographic c axis. The origin of the fishtail can be static, i.e., when theoretical $j_c(H)$ is a nonmonotonic function of field $H$, or it can be dynamic caused by field-dependent magnetic relaxation. Experimental determination of the origin of the fishtail in each material is, thus, very important as it allows shedding light on the nature of the flux pinning, hence, defect structure seen by the Abrikosov vortices. In Fe-based superconductors, interest is further fueled by multiple reports that defects, even nonmagnetic, are pair breaking, presumably, due to unconventional $s_\pm$ symmetry of the order parameter. 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 $H \rightarrow 0$. Therefore, to conduct a clean baseline experiment, one ideally needs a Fe-based superconductor with reduced scattering. These materials are rare but do exist in the form of only a few stoichiometric compounds, LiFeAs being one of them. Due to high sensitivity to air and moisture, there are only a few reports on the vortex properties in LiFeAs crystals. The fishtail effect and relatively high $j_B(5 \text{ K}) \approx 1 \times 10^5 \text{ A/cm}^2$ were found in Ref. 15, whereas, much lower $j_B(5 \text{ K}) \approx 1 \times 10^3 \text{ A/cm}^2$ was reported in Ref. 25. Such spread may be related to clean-limit superconductivity in this compound when even a small variation of impurity concentration causes significant change in the persistent current density and magnetic relaxation.

In this Rapid Communication, 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, the Labusch constant (and theoretical critical $j_c(H)$) is a monotonic function of the field when the Bean current (macroscopic vortex density gradient) is present, but it becomes a nonmonotonic function of the field at a homogeneous distribution of vortices. The values of $j_c(2 \text{ K}) \approx 1.22 \times 10^5 \text{ A/cm}^2$ provide an 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. At higher applied fields, vortex pinning and creep change to a collective regime 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 and were transported for measurements in sealed ampoules. Immediately after opening, $(0.5 - 1) \times (0.5 - 1) \times (0.1 - 0.3)\text{-mm}^3$ samples were placed into the cryostat for the measurements. Additionally, samples were extensively characterized by transport and magnetization measurements. Zero-field transition temperature of our samples was about $T_c \approx 18 \text{ K}$. The magnetic penetration depth was measured with the tunnel-diode resonator technique (for aza review, see Ref. 27). The sample was inserted into a 2-mm diameter copper coil that produced an rf excitation field (at $f \approx 14 \text{ MHz}$) of $H_{ac} \approx 20 \text{ mOe}$. An external dc magnetic field ($0 - 9 \text{ T}$) 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 cg units) is given by $\Delta f(T) = -G\pi\chi(T)$, 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 $R$, $4\pi\chi = (\lambda/L)\tanh(R/\lambda) - 1$, from which $\Delta\lambda$ can be obtained. The measured penetration depth consists of two terms, London penetration depth and Campbell penetration depth $\lambda_{Lm}^2 = \lambda_{L}^2 + \lambda_{C}^2$ (Ref. 17). Note that the measured penetration depth does not diverge at $T_c$ because it reaches the limiting value determined either by the size of the sample or by 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 $\lambda_{L}(0) = 200 \text{ nm}$ (Ref. 29).

Figure 1 shows the magnetic-penetration depth measured upon warming, after the sample was cooled in a zero field and the target field was applied at a low temperature (ZFC-W) compared to the measurements upon cooling when the target field was fixed above $T_c$ and was 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 $H = 250$-Oe field, which does not affect our analysis of the much higher fields. The inset in Fig. 1 shows an example of the small hysteresis of $\lambda_m(T)$ at $H = 7$ T.
indicating homogeneous vortex distribution]. The hysteresis between ZFC-W and FC-C-W is much smaller than, for example, that observed in Bi$_2$Sr$_2$CaCu$_2$O$_{8+y}$ crystals, which is most likely due to the much more three-dimensional 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 the zero field $\lambda_L(T)$ and the one measured in the 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 $j_c$ as a function of temperature at different magnetic fields $H_c(0) \approx 17$ T (Ref. 30), but $\xi \approx 7$ nm has been reported from the neutron-scattering form factor. Figure 3 shows $j_c$ as a function of temperature at different magnetic fields determined after the ZFC-W process (top frame) and the 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, reinforcing 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.

To understand the functional dependence, we plot determined $j_c(T)$ on a semilogarithmic 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 and can be well approximated by the exponential temperature dependence $j_c(1 \text{ T}) \approx 2.1 \exp(-T/3.1) \text{ MA/cm}^2$ for the FC-C process and $j_c(1 \text{ T}) \approx 2.3 \exp(-T/3.2) \text{ MA/cm}^2$ for the ZFC-W measurements. This very similar behavior implies 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, probably due to the 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 the theoretical critical current density $j_c$, determined from the ZFC Campbell penetration depth (top frame) and from the FC Campbell penetration depth (bottom frame) as a function of a 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 nonequilibrium ZFC process, is monotonic with the applied magnetic field at all temperatures, whereas, equilibrium $j_c$, obtained in the FC process when the 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.
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 therefore, we expect the true critical current density to be monotonic with the magnetic field. Since conventional (relaxed) dc measurements show the fishtail effect,\(^1\) we conclude that this effect is of dynamic origin. The functional form of the \( M(H) \) curve is governed by the nonmonotonic field-dependent pinning potential, implying the importance of vortex collective effects. More specifically, with the decrease in a magnetic field, the 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.\(^2\) It is possible that the origin of a fishtail in LiFeAs is similar to high-temperature cuprates. The question is how to reconcile very different (almost isotropic) electronic properties of Fe-based superconductors and quite similar to highly anisotropic cuprates vortex behavior.

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