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# Fishtail effect and vortex dynamics in LiFeAs single crystals

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We investigate the fishtail effect, critical current density ($J_c$), and vortex dynamics in LiFeAs single crystals. The sample exhibits a second peak (SP) in the magnetization loop only with the field || $c$ axis. We calculate a reasonably high $J_c$, however, values are lower than in Ba-122- and 1111-type FeAs compounds. Magnetic relaxation data imply a strong pinning which appears not to be due to conventional defects. Moreover, we find that the calculated magnetic relaxation rate is very low in LiFeAs. Our data suggest that the origin of the SP may be related to a vortex lattice phase transition. We have constructed the vortex phase diagram for LiFeAs on the field-temperature plane.

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## I. INTRODUCTION

The fishtail effect or the anomalous second peak (SP) in field ($H$) dependent magnetization ($M$) loops has been a subject of intense research in the field of superconductors with both low and high transition temperatures ($T_c$).[1–13] This phenomenon is realized with the enhanced irreversibility in isothermal $M(H)$ or, equivalently, the enhanced critical current density ($J_c$) at high fields apart from the central peak which occurs around zero field. In type-II superconductors, the magnetic fields above the lower critical field ($H_{c1}$) penetrate the bulk of the sample in the form of flux lines or vortices. Different mechanisms based on the vortex dynamics have been discussed to explain the SP in high-$T_c$ cuprates which include inhomogeneity of the sample,[2,3] matching effect,[4] surface barriers,[5] geometrical effects,[6] dynamic effects,[7] structural phase transition in the vortex lattice (VL),[8] vortex order-disorder phase transition,[9–12] crossover from elastic to plastic creep,[13] etc. However, in spite of plenty of studies dealing with this phenomenon, the general understanding lacks a converging trend and the proposed models appear to be more sample specific.

The recent discovery of superconductivity (SC) in Fe-based pnictides[14] has renewed the interest in vortex dynamics. Similar to cuprates, pnictides are also layer-based superconductors, and exhibit a high-$T_c$ and type-II nature. In contrast, pnictides have less anisotropy and larger coherence length ($\xi$), thus raising the question of how these influence the vortex dynamics in these materials. The appearance of a SP in $M(H)$ is not a universal phenomenon in different families of pnictides. For the 122 family ($A$Fe$_2$As$_2$, $A$ = Ba, Sr, Ca, etc.) the appearance of a SP is sensitive enough to the compositional elements. The pronounced SP has been observed, for example, in both hole- and electron-doped Ba-122 compounds, which has been ascribed to various mechanisms,[15–21] but it remains absent in doped Ca-122 compounds.[22] Similarly, a SP is not consistently seen in the 1111 ($R$FeAsO, $R$ = La, Nd, Ce, Sm, etc.) and in the 11 (FeTe) families.[23–25] However, the vortex dynamics have not been studied in great detail for the 111 family ($A$FeAs, $A$ = Li, Na, etc.).

Here we study the fishtail effect and the vortex dynamics in single crystals of LiFeAs which belong to the 111 family of pnictides. LiFeAs is an oxygen-free compound where superconducting active FeAs layers are separated by Li atoms along the $c$ axis.[26,27] Remarkably, LiFeAs exhibits SC in absence of any notable Fermi-surface nesting and static magnetism,[28] however, the presence of antiferromagnetic fluctuations in the normal state is inferred from the nuclear magnetic resonance (NMR) measurements.[29] Interestingly, recent experimental NMR results[30] and also theoretical calculations[31,32] indicate a possible $p$-wave SC state in LiFeAs, which is significantly different from other families within pnictides. LiFeAs is nonmagnetic and does not require any chemical doping to become superconducting, therefore the FeAs layers are more homogeneous and the crystal is devoid of coexisting magnetic phases, which makes it a good choice to study the vortex dynamics. Based on the sharpness of the rocking curve, recent small-angle neutron-scattering (SANS) measurements have revealed that the VL in LiFeAs exhibits no long-range order, however, better ordering than the doped Ba-122 compounds.[18,20,33]

We have investigated the properties of the vortex state in LiFeAs by means of isothermal $M(H)$ and magnetic relaxation measurements, which are the most extensively used tools for a variety of superconducting materials.[34,35] Our results imply a pronounced SP in both $M(H)$ and $J_c(H)$ at low temperatures ($T$) with the applied field parallel to the $c$ axis. We do not find a SP with the $H||ab$ plane. We determine the $J_c$ which is reasonably high, however, lower than those for doped Ba-122 and 1111 compounds. The magnetic relaxation data, on the other hand, imply a nonlogarithmic time ($t$) dependence with the estimated normalized magnetic relaxation rate ($S$) being very low (even lower than in Ba-122 and 1111 compounds) signifying a strong pinning for LiFeAs. Our data also indicate that the SP in LiFeAs may be due to a VL phase transition. From the $M(H)$ plots we have constructed the vortex phase diagram on the $H$-$T$ plane for LiFeAs.

## II. EXPERIMENTAL DETAILS

Single crystals of LiFeAs have been grown using the self-flux method as detailed in Ref. 36. The good quality and the homogeneity of the crystals are confirmed from the following experimental observations. (i) Analysis of the x-ray-diffraction (XRD) data using structural refinement implies the absence of any chemical impurity phase within the limit of experimental accuracy. (ii) The stoichiometry of the crystal has been confirmed by energy dispersive x-ray

(EDX) spectroscopy and by inductively coupled plasma mass spectroscopy (ICPMS) showing a molar ratio Li:Fe:As = 0.99:1:1. This result is quite remarkable because the deficiency in Li is known to produce different physical properties.[26] (iii) The local structural ordering of the crystal has been checked with nuclear quadrupole resonance (NQR). Usually, the NQR line becomes broadened due to inhomogeneities in charge distribution or lattice anomalies. Remarkably, the NQR signal of our crystal is very sharp with a linewidth (full width at half maximum) of 64 kHz, which is substantially smaller than in other pnictides.[37,38] (iv) We find a reasonably sharp transition in the magnetic susceptibility, resistivity, and specific-heat measurements.[39] Moreover, the residual resistivity shows a very low value of 0.025 mΩ cm favoring a good crystal quality. (v) An angle-resolved photoemission spectroscopy (ARPES) study performed on our crystal also confirms the stoichiometric composition and the uniform distribution of the Li content.[28]

For the present studies, two crystals ($S1$ and $S2$) of the same batch with rectangular shape have been selected. For the magnetic hysteresis loop sample $S1$ ($3.53 \times 2.5 \times 0.21$ mm$^3$) and for the magnetic relaxation measurements sample $S2$ ($2.89 \times 2.16 \times 0.38$ mm$^3$) have been used. Magnetization has been measured in a Quantum Design MPMS-XL superconducting quantum interference device. Adequate care has been taken to avoid the exposure of the sample to air before mounting it in the magnetometer. All the $M(H)$ and $M(t)$ measurements have been performed after cooling the sample in zero magnetic field from much above $T_c$ to the specific temperature. The $M(H)$ loops have been investigated with the field up to 50 kOe. For the relaxation measurements, the magnetization has been measured as function of time for about 8000 s.

## III. RESULTS AND DISCUSSIONS

The main panel of Fig. 1 presents the temperature dependence of the volume susceptibility ($\chi_{\mathrm{vol}}$) measured following the zero-field-cooled (ZFC) and field-cooled (FC) protocols for magnetization measurements. $\chi_{\mathrm{vol}}$ has been deduced from the measured dc $M$ with a field of 20 Oe applied parallel to the crystallographic $c$ axis. The data have been corrected for demagnetization effects.[40] It is evident from Fig. 1 that the sample exhibits bulk SC as characterized by the diamagnetic signal at low $T$. The sharp transition as well as the high value of $\chi_{\mathrm{vol}}$ in $M_{\mathrm{ZFC}}$ demonstrate the high quality of our crystal. We determine $T_c$ from the bifurcation point between ZFC and FC branches of the magnetization to be around 16.5(5) K. In the inset of the Fig. 1 we have plotted the $M(H)$ data at 10 K with $H$ parallel to both the $c$ axis and the $ab$ plane. With increasing $H$ for $H||c$, the magnetic irreversibility ($M_{\mathrm{irr}}$) initially decreases showing a minimum at a field $H_m$. On further increase in $H$, $M_{\mathrm{irr}}$ increases and exhibits a peak (SP) at a field $H_p$. However, we do not find any trace of a SP for the $H||ab$ plane. While this significant anisotropic behavior in the appearance of the SP is similar to other pnictide superconductors,[15] it remains different from cuprates, i.e., $\mathrm{La}_{1-x}\mathrm{Sr}_x\mathrm{CuO}_4$.[12]

Since the SP in $M(H)$ is only evident for $H||c$ in this compound, we have collected the $M(H||C)$ isotherms at different $T$ within the SC regime in order to understand the SP characteristics. The data are plotted in Fig. 2(a) where $M$ vs $H$ loops are quite symmetric with respect to both the sweeping direction as well as the polarity of the magnetic field. At low $T$, however, even though the onset of the SP is evident, the SP cannot be observed within the measurable field range. Interestingly, $M(H)$ at 2 K exhibits irregular jumps close to $H = 0$. These jumps are commonly known as flux jump effects[41] and will be presented elsewhere in more detail. With the increase in $T$, a clear SP can be observed in the $M(H)$

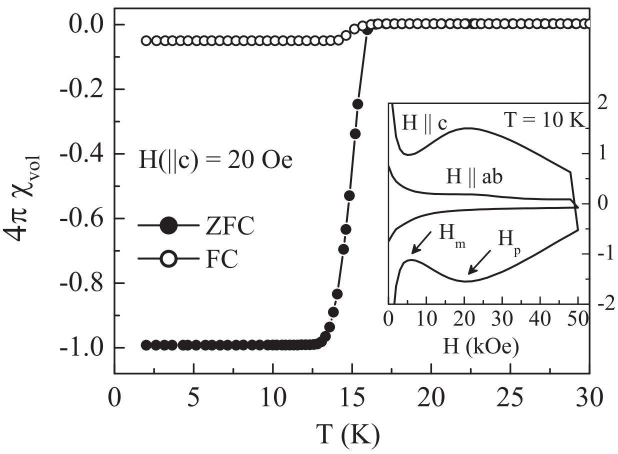

FIG. 1. Temperature dependence of $\chi_{\mathrm{vol}}$ for LiFeAs deduced from dc $M$ measured following ZFC and FC protocols. The data have been collected in $H||c = 20$ Oe. The inset shows $M$ vs $H$ plots measured at 10 K with the field parallel to both $c$ axis and $ab$ plane.

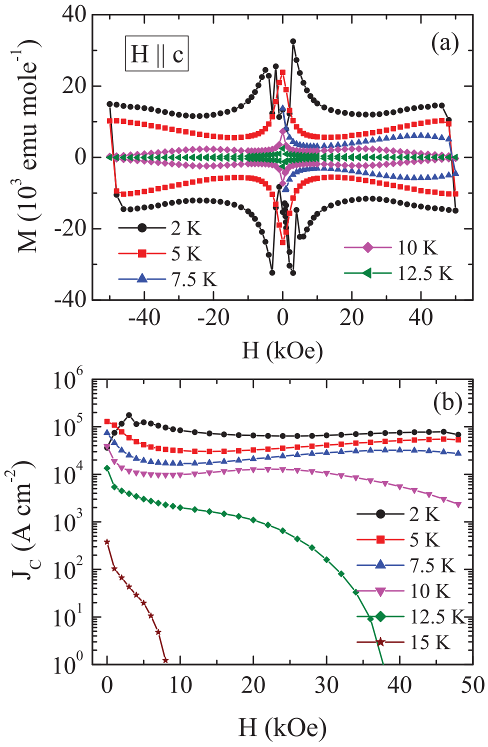

FIG. 2. (Color online) (a) The isothermal $M$ vs $H$ loops recorded at different temperatures with $H||c$ for LiFeAs. (b) The critical current density $J_c$ as calculated from the $M(H)$ loops in (a) as function of field for different temperatures; for details see text.

loops. Moreover, we find a field $H_{\rm irr}$ above which $M_{\rm irr}$ in data vanishes. With increasing $T$, all the characteristics fields, i.e., $H_m$, $H_p$, and $H_{\rm irr}$ decrease, and their $T$ variation will be discussed in a later section.

From the magnetic irreversibility in $M(H)$ we have calculated the critical current $J_c$ exploiting the Bean's critical state model[42] $J_c = 20\Delta M/[a\,(1 - a/3b)]$, where $\Delta M = M_{\rm dn} - M_{\rm up}, M_{\rm up}$, and $M_{\rm dn}$ are the magnetization measured with increasing and decreasing field, respectively, and $a$ and $b$ $(b > a)$ are the dimensions of the crystal perpendicular to the applied $H$. The unit of $\Delta M$ is in emu/cm$^3$, $a$ and $b$ are in cm, and the calculated $J_c$ is in A/cm$^2$. The calculated $J_c(H)$ has been plotted in Fig. 2(b) for different $T$. The variation in $J_c(H)$ is nonmonotonic and exhibits a broad peak (SP) in the high-field region, which is in line with $\Delta M$ in Fig. 2(a). At low $T$, $J_c$ is rather high, however, its value still being lower than in doped Ba-122 and 1111 compounds where $J_c \sim 10^6$ A/cm$^2$ or even higher.[15–17,19,24] This is in agreement with the level of disorder as revealed from the SANS studies.[18,20,33]

To understand the origin of the SP and the reasonably high $J_c$ values in LiFeAs, we have studied detailed vortex dynamics in this compound by means of $T$- and $H$-dependent magnetic relaxation measurements.[34,35] Magnetic relaxation in superconductors is a result of nonequilibrium spatial arrangement of vortices due to pinning sites. External applied magnetic field exerts Lorentz forces on the vortices resulting in their movement, which causes a change in $M(t)$. In contrast to the original Anderson-Kim model,[43] our relaxation data exhibit a nonlogarithmic time dependence and can be best fitted with the interpolation formula:[34]

$$M(t) = M_0\left[1 + \frac{\mu k_{\rm B}T}{U_0}\ln\left(\frac{t}{t_0}\right)\right]^{-1/\mu}, \qquad (1)$$

where $k_B$ is the Boltzmann constant, $U_0$ is the energy barrier height in absence of a driving force, $t_0$ is the characteristic relaxation time (usually $\sim 10^{-6}$ s for type-II superconductors), and $\mu$ is the field-temperature-dependent parameter. This formula yields the normalized magnetic relaxation rate $S[= (1/M)dM/d\ln(t)]$ as[34]

$$S(t) = \frac{k_{\rm B}T}{U_0 + \mu k_{\rm B}T\ln(t/t_0)}. \qquad (2)$$

Magnetic relaxation has been measured at different $T$ in $H||c = 10$ kOe. We find a very slow relaxation, i.e., there is only a 4% change in magnetic moment at $H = 10$ kOe and $T = 10$ K within a time period of 8000 s which is much lower than that observed in cuprates and 122-pnictides.[15,34] $U_0/k_B(T)$ and $S(T)$ as extracted from the fitting of the data exploiting Eqs. (1) and (2) are plotted in Figs. 3(a) and 3(b), respectively. One representative fitting of our data has been included as an inset in Fig. 3(b). Surprisingly, we find a very high value of $t_0$ of the order of 10 s, which is orders of magnitude higher than for other families in pnictides and cuprates.[15,22,34,35] For LiFeAs the value for the energy barrier $U_0/k_B$ increases with $T$, however, above 10 K it drops down due to larger thermal fluctuations. A similar behavior can be detected for $S(T)$, which also shows a nonmonotonous behavior (calculated at $t = 1000$ s) with a steep decrease followed by an increase upon lowering temperature.

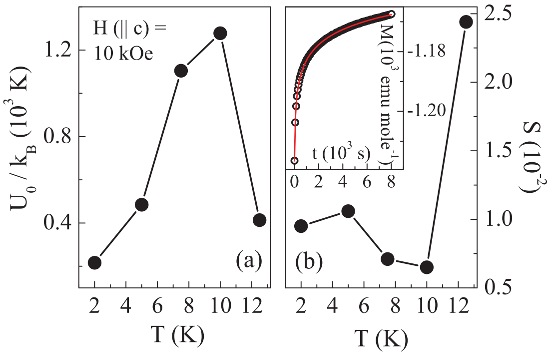

FIG. 3. (Color online) (a) The energy barrier $U_0/k_B$ obtained by fitting of Eq. (1) as a function of temperature. (b) The magnetic relaxation rate $S$ as a function of temperature in $H||c = 10$ kOe as calculated using Eq. (2) at $t = 1000$ s. The inset exemplary shows the best fit of the magnetic relaxation data at $T = 10$ K and $H||c = 10$ kOe using Eq. (1).

In addition to the temperature-dependent relaxation studies, $M(t)$ has been measured in different fields $(H||c)$ along the hysteresis loop at constant $T$. Similarly, we have extracted $U_0/k_B$ and $S$ using Eqs. (1) and (2). Figure 4(a) presents the results for $U_0/k_B(H)$ and $S(H)$ at 10 K. Here, the vertical dashed lines in Fig. 4(a) represent the minimum and maximum (SP) in $M(H)$ at 10 K (see Figs. 1 and 2). It is evident from the plot that $U_0/k_B$ initially increases with applied fields and then decreases, showing a peak at a field between $H_m$ and $H_p$, where the related calculated $S(H)$ exhibits a minimum. This variation in $S(H)$ is similar to doped Ba-122 compounds.[15,21] To examine the behavior of $U_0/k_B(H)$ and $S(H)$ at low $T$ where the SP shifts significantly to higher field values, we have calculated the parameters at 5 K following the same method. Our results show that although the field, where $U_0/k_B$ $(S)$ exhibits a maximum (minimum), increases from ≈10 kOe at 10 K to ≈20 kOe at 5 K [Fig. 4(b)], this variation does not scale with the large increase in $H_p$ at low $T$ [see Fig. 2(a)]. Note that the calculated $S$ in Figs. 3 and 4 is very low, and that the values even go below those for Ba-122 and 1111 compounds, where $S > 0.01$.[15,21,23,24] The low value of $S$ paired with our very high $t_0$ value imply a high pinning in LiFeAs. This is quite intriguing as $J_c$ in LiFeAs is lower than in Ba-122 and 1111

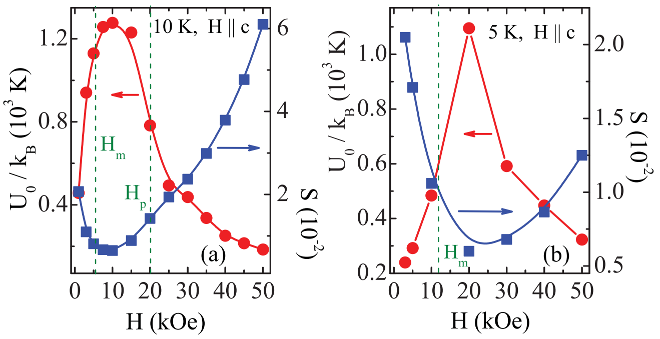

FIG. 4. (Color online) (a) The field dependence of $U_0/k_B$ and $S$ (at 1000 s) extracted from the magnetic relaxation data at 10 K exploiting Eqs. (1) and (2), respectively. The vertical dashed lines mark $H_m$ and $H_p$; for details see text. (b) The same physical properties $U_0/k_B$ and $S$ shown for 5 K.

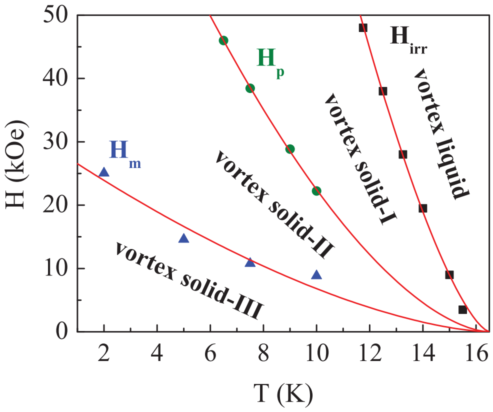

FIG. 5. (Color online) The vortex phase diagram on the $H$-$T$ plane with $H||c$ for LiFeAs. Solid lines represent the fit to the experimental data using the functional form $H_x(T) = H_x(0)(1 - T/T_c)^n$ (see text).

materials,[15–17,19,24] which means that other types of pinnings rather than the conventional defects are active in LiFeAs. Indeed, our crystal is of good quality and homogeneous as discussed earlier. Therefore further detailed investigations are required to understand this interesting pinning behavior in LiFeAs.

From the so-far-obtained data we have constructed the vortex phase diagram on the $H$-$T$ plane for LiFeAs (Fig. 5). Above $H_{\text{irr}}$, vortices are in a liquid state. Below $H_{\text{irr}}$, they are in a solid state, however, its nature changes between different field regimes indicated in the figure (named I, II, and III). All the characteristic fields show a strong $T$ dependence where the data can be fitted well with the functional form $H_x(T) = H_x(0)(1 - T/T_c)^n$. We obtain $H_{\text{irr}}(0) = 299.5(9)$ kOe and $n = 1.46(4)$, $H_p(0) = 105.7(8)$ kOe and $n = 1.66(3)$, $H_m(0) = 29.1(4)$ kOe and $n = 1.56(3)$. The values of the exponent $n$ are reasonably consistent with those for other pnictides.[15,16,23,24]

Now we discuss the origin of the SP effect and the intriguing vortex dynamics in LiFeAs. The strong $T$ dependence of the transition lines (Fig. 5) discards the possibility that the SP in LiFeAs arises due to a vortex order-disorder phase transition.[9–12] Moreover, an absence of "mirror-image" correlation between $M(H)$ and $S(H)$ in Figs. 2 and 4 implies that the dynamic model[7] is not valid in the present case. Although the nature of $U_0/k_B(H)$ in Fig. 4 has qualitative similarity to the model which predicts the SP being associated with a crossover in flux dynamics from elastic ($< H_p$) to plastic ($> H_p$) creep with increasing field,[13] the peak in $U_0/k_B(H)$ for LiFeAs occurs much below the SP, and in low $T$ at 5 K this mismatch increases. Moreover, this model predicts $H_p \propto [1 - (T/T_c)^4]^{1.4}$, which does not describe the present $H_p(T)$ dependence shown in Fig. 5. Henceforth the validity of this model for LiFeAs is questionable.

On the other hand, a structural phase transition in the VL is also an attractive and possible model which argues that the SP is associated with the transformation of a hexagonal VL to a square one with field.[8] The square structure is supported by the fourfold symmetry of the intervortex interaction which can originate in various situations, like, for the anisotropic ($d$-wave) nature of the superconducting gap as in $La_{2-x}Sr_xCuO_4$,[44] for materials with low Ginzburg-Landau (GL) parameters ($\kappa$) as in $YNi_2B_2C$,[45] in the extended GL theories with more than one order parameter as for the $p$-wave superconductor $Sr_2RuO_4$,[46] etc. Recently, such scenario of a structural phase transition in the VL has been proposed in $Ba(Fe_{0.925}Co_{0.075})_2As_2$ where a minimum in $S(T)$ and $S(H)$ has been found.[21] Altogether, taking into account a similar behavior with a minimum in $S(T)$ and $S(H)$ in LiFeAs, paired with a comparatively low value of $\kappa$ ($\approx$30) (Ref. 33) and the proposed $p$-wave SC (Refs. 30–32) in LiFeAs, a structural phase transition in the VL is a possible scenario for the existence of the SP in this compound. However, further investigations including microscopic probes are required to confirm these observations.

## IV. CONCLUSION

In summary, single crystalline LiFeAs exhibits a SP in the $M(H)$ loop with $H||c$ axis. The calculated $J_c$s are reasonably high, however, the values are lower than in the doped Ba-122 and 1111 compounds. We find an extraordinary slow magnetic relaxation implying a strong pinning which appears not to be related to conventional defects. We have constructed the vortex phase diagram on the $H$-$T$ plane for LiFeAs, with the characterized fields $H_{\text{irr}}$, $H_p$, and $H_m$ showing a strong $T$ dependence. In accordance with recent investigations on $Ba(Fe_{0.925}Co_{0.075})_2As_2$ our data imply that the SP in LiFeAs most likely originates from a VL phase transition. Nonetheless, further studies involving microscopic probes are required to comprehend the SP and vortex dynamics in this compound.

## ACKNOWLEDGMENTS

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