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Upper critical field and thermally activated flux flow in $\text{LaFeAsO}_{1-x}\text{F}_x$

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The magneto-resistance, critical current density, J_c , upper critical field, H_{c2} , and flux pinning properties of $\text{LaFeAsO}_{1-x}\text{F}_x$ superconductors were investigated systematically by magnetic and magneto-transport measurements in the fields up to 13 T over a temperature range of 5–35 K. It was found that the H_{c2} increased with increasing fluorine concentration up to $x \leq 0.15$, while with higher fluorine doping, H_{c2} decreased. A peak effect in the J_c as a function of field was observed at $T < 15$ K for both the 5% and 15% fluorine doped samples. The broadening of the superconducting transition in magnetic field can be well understood by the thermally activated flux flow model. The pinning potential, U_ϕ , scales as $U_\phi/k_B \propto B^{-n}$ with $n = 0.13$ for $B < 1$ T and $n = -0.68$ for $B > 1$ T for $\text{LaFeAsO}_{0.85}\text{F}_{0.15}$. © 2011 American Institute of Physics. [doi:10.1063/1.3566069]

For practical application of the Fe-based superconductors, two of the most important parameters are H_{c2} , and J_c . The H_{c2} is an intrinsic property, which has been estimated to be higher than 64 T in $\text{LaFeAsO}_{0.89}\text{F}_{0.11}$,¹ over 100 T in $\text{SmFeAsO}_{0.85}\text{F}_{0.15}$,² and 230 T in $\text{NdFeAsO}_{0.82}\text{F}_{0.18}$.³ However, J_c is sample dependent and determined by pinning potential and vortex behavior at high field and temperature. Critical current density and flux pinning behavior have been well studied for $\text{PrFeAsO}_{0.9}$,⁴ $\text{NdFeAsO}_{1-x}\text{F}_x$,^{3–7} and $\text{SmFeAsO}_{0.7}\text{F}_{0.25}$ (Ref. 8) single crystals.

As La is a nonmagnetic element, the $\text{LaFeAsO}_{1-x}\text{F}_x$ compounds are ideal candidates for the study of the flux pinning related properties. This is because all the other RE-1111 compounds contain a magnetic RE. In this work, we report on the effects of different fluorine concentrations on the upper critical field, critical current density, and pinning potentials in the $\text{LaFeAsO}_{1-x}\text{F}_x$ polycrystalline samples.

Polycrystalline samples with the nominal composition of $\text{LaFeAsO}_{1-x}\text{F}_x$ ($x = 0.05, 0.15$, and 0.20) were prepared by a conventional solid state reaction. The details of the preparation process can be found elsewhere.⁹ X-ray diffraction (XRD) experiments were performed with the Cu-K α radiation in the 2θ range from 20° to 80° , with a step interval of 0.02° . The crystal structures were refined using Rietveld refinement. The transport and magnetic properties were measured over a wide range of temperature and magnetic fields up to 13 T, using a physical properties measurement system (PPMS, Quantum Design).

A typical x-ray diffraction pattern and refinement results of a $\text{LaFeAsO}_{1-x}\text{F}_x$ ($x = 0.05$) sample are shown in Fig. 1.

These can be seen that the as-prepared sample is nearly single phase with tetragonal structure having the P4/nmm symmetry and the lattice parameters $a = 4.023$ Å and $c = 8.709$ Å. The J_c values were calculated from the M-H loops at different temperatures by using the extended Bean model:

$$J_c = 20 \Delta M / [l(1 - l/3w)], \text{ with } l < w,$$

where l and w are the sample dimensions perpendicular to the applied field, and ΔM is the height of the magnetization loop. The J_c versus magnetic field at different temperatures are shown in Fig. 2. At 5 K for the $x = 0.15$ sample, the J_c is approximately 10^3 A/cm² at zero field and then decreases to 320 A/cm² at 1.2 T. The J_c increases slightly with increasing magnetic field and reaches a peak value of 330 A/cm² at about 2.2 T (Fig. 2), then decreases with field. The same peak effect can be seen at 10 and 15 K, but in lower magnetic fields. A similar peak effect has also been detected for $\text{LaFeAsO}_{0.95}\text{F}_{0.05}$. The observation of the peak effect in our La-1111 compounds is in agreement with what has been reported in $\text{SmFeAsO}_{1-x}\text{F}_x$ (Ref. 2) and $\text{NdFeAsO}_{1-x}\text{F}_x$ (Ref. 10) compounds.

The temperature dependence of resistance for samples with different fluorine concentrations at zero magnetic field is shown in the inset of Fig. 3. The resistance starts to drop at the temperature of 27.5, 26, and 27.5 K and then vanishes below 25.5, 24, and 25.5 K for the 5%, 15%, and 20% fluorine doped samples, respectively. The resistance for the $x = 0.05$ and 0.2 fluorine doped samples decreases linearly with decreasing temperature down to T_c , whereas, the resistance for the $x = 0.15$ sample decreases down to 70 K and then increases again, reaching a maximum at the onset of T_c . A similar resistance peak anomaly has been detected in other superconductors and it is usually related to sample

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FIG. 1. (Color online) Observed (symbols) and calculated (solid line) XRD patterns of $\text{LaFeAsO}_{1-x}\text{F}_x$ ($x=0.05$) with the bottom trace showing the difference. The blue lines indicate the peak positions from the La1111 phase. The inset shows the crystal structure of the $\text{LaFeAsO}_{1-x}\text{F}_x$.

inhomogeneities and enhanced electron scattering in the vicinity of T_c .^{11–13} The residual resistance ratio ($\text{RRR} = R_{300\text{ K}}/R_{2\text{ K}}$) is 7.98, 2.67, and 8.72 for the 5%, 15%, and 20% fluorine doped samples, respectively, which indicates that the scattering for the 5% and 20% fluorine doped samples is higher than for the 15% fluorine doped sample. The $\Delta T_c = T_c(90\%) - T_c(10\%)$ for zero magnetic field is only 2.4, 1.8, and 2.6 K for the 5%, 15%, and 20% fluorine doped samples, respectively, which is much smaller than the value of 4.5 K for $\text{LaFeAsO}_{0.89}\text{F}_{0.11}$ reported earlier,¹⁴ indicative of relative good chemical homogeneity of our samples.

As the J_c drops to zero at a temperature of about 20 K in $H > 2$ T, the 90% and 10% of normal state resistance can be regarded as the upper critical field in the ab -plane and along c direction, H_{c2}^{ab} and H_{c2}^c , respectively.¹ The temperature dependence of upper critical field in ab plane and along c direction is shown in Fig. 4. The estimated slope dH_{c2}^{ab}/dT for H_{max} is -4.1 , -6.8 , and -3.5 TK^{-1} for 5%, 15%, and 20% fluorine doped samples, respectively which are greater than those reported for $\text{LaFeAsO}_{0.89}\text{F}_{0.11}$ ($dH_{c2}/dT = 2$ TK^{-1}) and for $\text{NdFeAsO}_{0.82}\text{F}_{0.18}$ ($dH_{c2}/dT = 5.8$ TK^{-1}).³ The dH_{c2}^{ab}/dT for H_{min} (10% R_n) is -1.8 , -2.1 , and -1.5 T

FIG. 2. (Color online) Magnetic field dependence of the critical current density at different temperatures for the three samples.

FIG. 3. (Color online) The temperature dependence of the resistance of $\text{LaFeAsO}_{1-x}\text{F}_x$ samples at different magnetic field. The inset shows the temperature dependence of resistance for $\text{LaFeAsO}_{1-x}\text{F}_x$ at zero magnetic fields.

K^{-1} for the 5%, 15%, and 20% fluorine doped samples, respectively. The estimated $H_{c2}^{ab}(0)$ using the Werthamer–Helfand–Hohenberg (WHH) formula: $H_{c2}(0) = -0.693 T_c (dH_{c2}/dT)_{T=T_c}$, are 78, 122, and 69 T along ab plane and $H_{c2}^c(0)$ is 34.3, 37.8, and 29 T for the 10% R_n fields for the 5%, 15%, and 20% fluorine doped samples, respectively. These values are much greater than the highest value of 60 T which was reported earlier.³

It has been reported that the Pauli-limiting (PL) effect is responsible for the H_{c2} values in high fields in As-deficient $\text{LaFeAs}_{1-y}\text{O}_{0.9}\text{F}_{0.1}$ superconductors. The H_{c2} due to the PL effect is lower than what is estimated using the WHH formula and can be calculated using $B_{c2}^p(0) = B_{c2}^*(0)/\sqrt{1+\alpha^2}$, where α is the Maki parameter. The α values are sample dependent and can vary between 0.25 and 3 for the La-1111 and Nd-1111 compounds.¹⁵ α is 1.31 for As-deficient $\text{LaFeAs}_{1-y}\text{O}_{0.9}\text{F}_{0.1}$.¹⁶ As our samples are not As deficient, it should have a small value of $\alpha = 0.25$ as reported for La-1111 samples. Therefore, the $B_{c2}^p(0)$ values should be almost the same as those obtained from the WHH estimation. It should be noted that we are unable to estimate α values precisely for our samples because of the lack of H_{c2} data in high magnetic fields. The Ginzburg–Landau coherence length at zero temperature ($\xi_{\text{GL}} = (\phi_0/2\pi$

FIG. 4. (Color online) Upper critical field—temperature phase diagram of $\text{LaFeAsO}_{1-x}\text{F}_x$.

FIG. 5. (Color online) (a) Arrhenius plot of the electrical resistance of $\text{LaFeAsO}_{0.95}\text{F}_{0.05}$. U_0 is given by the slope from a linear fitting. (b) Magnetic field dependence of U_0 for $\text{LaFeAsO}_{1-x}\text{F}_x$ ($x = 0.05, 0.15, 0.20$).

$H_{c2}^{\text{ab}})^{1/2}$, where $\varphi_0 = 2.07 \times 10^{-7} \text{ Oe} \cdot \text{cm}^2$) is 2.06, 1.64, and 2.18 nm for the 5%, 15%, and 20% fluorine doped samples, respectively. These values are smaller than the reported values for $\text{LaFeAsO}_{0.89}\text{F}_{0.1}$ compound.¹⁴ Furthermore, the estimated anisotropy γ ($=H_{c2}^{\text{ab}}/H_{c2}^{\text{c}}$) in the temperature range of $17 < T < 28 \text{ K}$ is 2.3, 3.2, and 2.4 for the 5%, 15%, and 20% fluorine doped samples, respectively.

The broadening of the superconducting transition in the magnetic field is caused by the thermally activated flux flow in $\text{NdFeAsO}_{1-x}\text{F}_x$ superconductor.⁵ Thus we use the same model to discuss the broadening of resistance in our samples. According to the thermally activated flux flow model, the resistance is $R(T, B) = R_0 \exp[-U_0/k_B T]$, where U_0 is the flux-flow activation energy, which can be obtained from the slope of the linear part of an Arrhenius plot, R_0 is a parameter, and k_B is Boltzmann's constant. In Fig. 5, we plot the data as $\ln R$ versus T^{-1} . The thermally activated behavior of the resistance is immediately apparent. The best fit to the experimental data yields values of the activation energy, $U_0/k_B = 1130$ and 910 K in the low field of 0.1 T for the 5% and 15% fluorine doped samples, respectively. These values are lower than U_0 value for a polycrystalline $\text{NdFeAsO}_{0.82}\text{F}_{0.18}$ sample ($U_0/k_B = 2000 \text{ K}$ for a magnetic field of 0.1 T).³

Figure 5(b) shows the magnetic field dependence (up to 13 T) of U_0 for all samples. We can see that the values of U_0 for $\text{LaFeAsO}_{0.85}\text{F}_{0.15}$ drop very weakly with field for $B < 1 \text{ T}$, scale as $B^{-0.13}$, and then decrease as $B^{-0.68}$ for $B > 1 \text{ T}$. The activation energy of the 5% and 20% fluorine doped samples shows a similar trend with field. The field dependence of U_0 in our samples is the same as what we have found for $\text{NdFeAsO}_{1-x}\text{F}_x$ sample, indicating that the vortices

should have the same nature in both La-1111 and the Nd-1111 polycrystalline superconductors. As the U_0 decreases weakly with field, it is expected that single-vortex pinning may co-exist with collective creep in low field ($< 1 \text{ T}$), and then the collective creep dominates for $B > 1 \text{ T}$ for all our La-1111 samples with various F concentrations.

In summary, $\text{LaFeAsO}_{1-x}\text{F}_x$ with various F concentrations shows a superior J_c field dependence at low temperatures. A peak effect in the J_c versus field was observed in the $x = 0.05$ and 0.15 samples. High value of $dH_{c2}^{\text{ab}}/dT = 6.8 \text{ T K}^{-1}$ was obtained for the $x = 0.15$ sample with the $H_{c2}^{\text{ab}}(0) = 122 \text{ T}$. The thermally activated flux flow is responsible for vortex behaviors in the tails of the RT cures.

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