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Enhanced upper critical field, critical current density, and thermal activation energy in new ytterbium doped $CeFeAsO_{0.9}F_{0.1}$ superconductor

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In this report, we have investigated the essential physical properties of $Ce_{0.7}Yb_{0.3}FeAsO_{0.9}F_{0.1}$ superconductor such as field dependent critical current density (J_c), thermal activation energy (U_0), and upper critical field (H_{c2}). From the isothermal magnetization curves and size of the superconducting grains, the critical current density J_c of 2.3×10^6 A/cm^2 at 2 K, 0.5 T was estimated using the Bean's model for this Yb doped superconductor. A gradual decrease of J_c and absence of peak effect were found on increasing magnetic field up to 5 T. Thermal activation energy ($U_0/k_B = \sim 2500$ K) calculated from Arrhenius plots at low magnetic field (0.5 T) indicates a strong flux pinning potential might be co-existing with applied magnetic field. Our results suggest that this new Yb doped superconductor is a possible practical high temperature superconductor under certain magnetic field and temperature. © *2013 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4788799]

I. INTRODUCTION

The interest and importance of any superconductors rely on how these superconductors and devices based on them work at high temperature with maximum sustainability. Among the many essential properties of superconductors, the upper critical field (H_{c2}) and critical current density (J_c) are two predominant parameters that decide the practicability of the material. For polycrystalline samples, a weak intergranular coupling of grains and fluctuation in the grain boundaries under magnetic field restrict the flow of supercurrent, and their use as superconducting magnets. New superconductors are needed in order to investigate these parameters and extend the applicability of superconducting hybrid devices. It has been more than 5 yr since the first iron arsenide superconductors were found by Kamihara *et al.*[1] After that many compounds with a similar two dimensional layered structure have been reported[2–4] with maximum T_c of $\sim$57 K in $Ca_{0.4}Nd_{0.6}FeAsF$ and $Sm_{0.95}La_{0.05}FeAsO_{0.85}F_{0.15}$.[5,6]

It should be noted that due to a larger anisotropy and imperfection in the microstructure of superconducting polycrystalline grains, the flux pinning potential and critical current density are expected to show lower values. The upper critical field of $\sim$46 T, with a T_c (onset) of about $\sim$29 K was found in $LaFeAsO_{0.9}F_{0.1}$ (Ref. 7) and both H_{c2} (0) and T_c could be enhanced up to $\sim$150, 204 T, and $\sim$46 K, respectively, on replacing La by Sm and Nd.[8,9] Several investigations have already shown that the value of J_c (A/cm^2) is higher under very low magnetic field and with high transition temperature (T_c). The studies on field dependence of J_c for $SmFeAsO_{0.7}F_{0.3}$ and $NdFeAsO_{0.7}$ polycrystalline samples estimate a J_c of about 5×10^5 A/cm^2 at very low magnetic field,[10] which is smaller than that of YBCO single crystals.[11]

Zhigadlo *et al.* and Yang *et al.* reported that the single crystals of $SmFeAsO_{1-x}F_x$ and $B_{0.6}K_{0.4}Fe_2As_2$ have capability of carrying supercurrent of the order of 10^6 A/cm^2, which was the maximum J_c observed so far at very low temperatures in FeAs based compounds.[12,13] Perhaps, defect-free single crystals would show higher critical current rather than polycrystalline samples due its homogeneous crystalline structure. In this paper, we have analyzed and compared the relation between upper critical field, micro-crystalline structure, critical current density, and thermal activation energy of this polycrystalline sample by using Werthamer-Helfand-Hohenberg (WHH) formula, scanning electron microscopic (SEM) studies, Beans model, and Arrhenius plots.

II. EXPERIMENT

The polycrystalline sample with nominal composition $Ce_{0.7}Yb_{0.3}O_{0.9}F_{0.1}FeAs$ was synthesized using two step solid state reaction method and structural properties were analyzed by powder X-ray diffraction.[14] The M vs H hysteresis loops below T_c were measured by Physical Property Measurement System (PPMS) - Vibrating Sample Magnetometer (VSM) (Quantum Design, USA). Microstructure analyses of polycrystalline grains were carried out using SEM operated at electron energy of 20 kV. The field dependent critical current density (J_c) was calculated using the Bean's model. Thermal activation energy (U_o) was calculated from the slope of the linear temperature dependent Arrhenius plots. $Ce_{0.7}Yb_{0.3}FeAsO_{0.9}F_{0.1}$ showed a T_c ($\sim$42 K)[14] higher than the parent $CeFeAsO_{0.9}F_{0.1}$ ($T_c \sim 38$ K) due to a increase in chemical pressure brought by substitution of smaller Yb atoms in Ce site.[15] From the resistivity measurements, the calculated residual resistivity value ($RRR = R_{300K}/R_{50K}$) above 50 K was found to be

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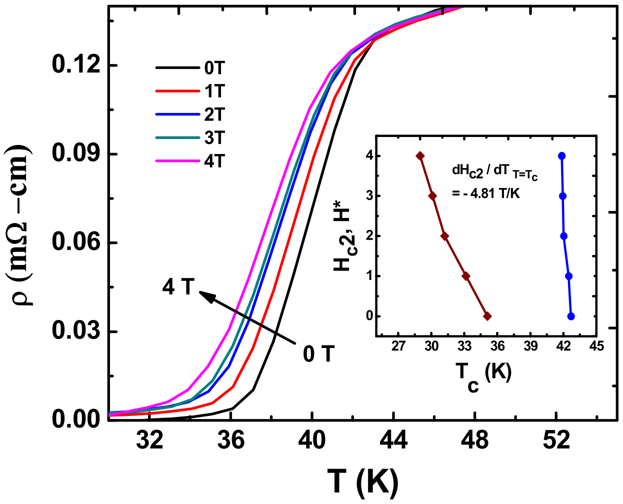

FIG. 1. The magnetic field dependence of resistivity as a function of temperature for $Ce_{0.7}Yb_{0.3}FeAsO_{0.9}F_{0.1}$. Inset shows temperature dependence of upper critical field (•) and irreversibility field (◆).

3.6, which is close to the values of $CeFeAsO_{0.8}F_{0.2}$ and $Ce_{0.6}Y_{0.4}FeAsO_{0.8}F_{0.2}$ reported by Prakash *et al.*[16]

III. RESULTS AND DISCUSSIONS

Figure 1 shows the magnetoresistance plots for $Ce_{0.7}Yb_{0.3}FeAO_{0.9}F_{0.1}$ as a function of temperature, which gives the details of upper critical field (H_{c2}) and flux pinning properties at high magnetic field. It can be seen that the T_c shifts to lower temperature on increasing the applied magnetic field. The upper critical field (H_{c2}) and irreversibility field (H^*) were obtained using the 90% and 10% values of normal state resistivity (ρ_n) curves and its relation to the transition temperature is shown in the inset of Fig. 1. The upper critical field at 0 K was estimated using WHH formula[17] ($H_{c2}(0) = -0.693\ T_c\ (dH_{c2}/dT)_{T=Tc}$). Using H_{c2} versus T plot (inset of Fig. 1), the slope (dH_{c2}/dT) was calculated to be −4.81 [T/K]. $H_{c2}(0)$ was found to be ∼142 T for $Ce_{0.7}Yb_{0.3}FeAsO_{0.9}F_{0.1}$ ($T_c = 42.6$ K), which is higher than ytterbium free $CeFeAsO_{0.9}F_{0.1}$ superconductor (∼94 T).[15] In order to study the microstructure and dispersion of superconducting grains in this polycrystalline compound, we have investigated the SEM images of $Ce_{0.7}Yb_{0.3}FeAsO_{0.9}F_{0.1}$ (Figs. 2(a) and 2(b)). The particles appeared to be agglomerated and rough estimation of grain size yielded size within 10 and 20 μm range.

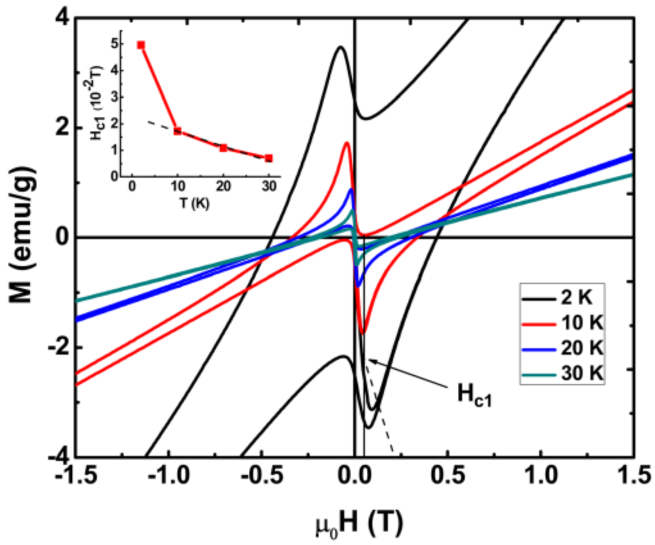

FIG. 3. Magnetic hysteresis loops for $Ce_{0.7}Yb_{0.3}FeAsO_{0.9}F_{0.1}$ measured at 2 K, 10 K, 20 K, and 30 K. Arrow marks indicate the methodology of observed lower critical field (H_{c1}). Inset illustrates H_{c1} as a function of temperature plot.

The field dependence of magnetization was also studied at different temperatures (2 K, 10 K, 20 K, and 30 K) as shown in Fig. 3. It is clear that the observed hysteresis loops for all temperatures indicate the presence of small Fe based magnetic impurities such as FeAs, which is also confirmed by X-ray diffraction analysis.[14] The lower critical field (H_{c1}) can be defined as the deviation from the linear M(H) curve as shown in the same figure. H_{c1} is found to be around 500 Oe at 2 K and it decreases to 70 Oe on increasing the temperature from 2 to 30 K. Inset of Fig. 3 shows that H_{c1}(T) slightly deviates from straight line, which may be due to possible suppression of flux flow at lower temperatures. Figure 4 shows the magnetic field dependence of critical current density at 2 to 30 K. The intragrain current density (J_c) was calculated using the Bean's model[18] from M (B) curves ($J_c = 30\ \Delta M/d$, where ΔM is the difference in decreasing and increasing branch of magnetization (($M_{dec} - M_{inc}$)/2) under

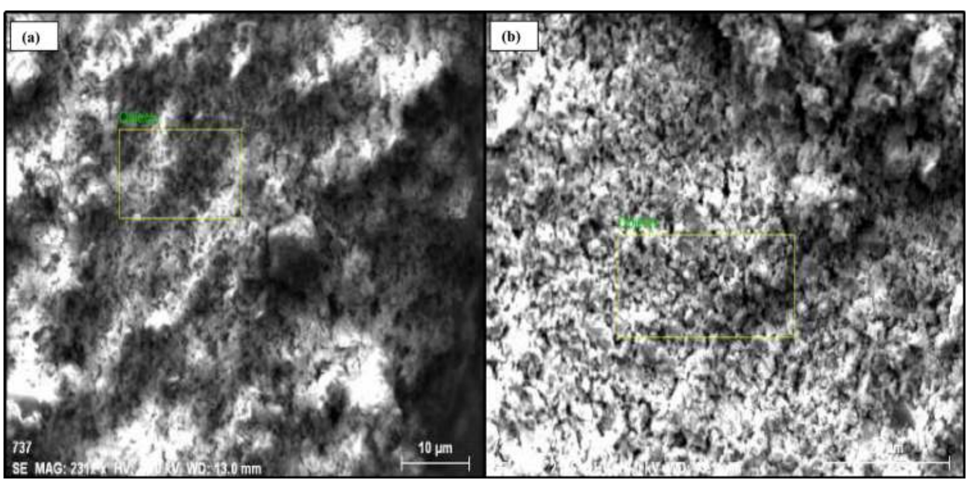

FIG. 2. SEM micrographs of superconducting grains within the ranges of (a) 10 and (b) 20 μm.

applied magnetic field (in Gauss) and d is the average grain size (15 μm). Using the Bean's model, the calculated J_c value was found to be 2.3×10^6 A/cm^2 at 2 K at low magnetic field (0.5 T). A gradual decrease in J_c was found on increasing magnetic field (B > 3 T) and there is no indication of any peak effect up to a magnetic field of 5 T,[19] which is consistent with the change of J_c under low field for Nd and Sm based oxypnictides.[10] As shown in Fig. 4, J_c decreases from 2.3×10^6 A/cm^2 to 3.7×10^3 A/cm^2 within the temperature range of 2 to 30 K and field range of 0.5 T to 5 T suggesting slightly weak links between the grains, which are also confirmed by the presence of small hysteresis loops in magnetization studies (Fig. 3). Inset of Fig. 4 depicts that as the temperature increases critical current density shifts to lower values, which is in good agreement with previously reported superconductors.[11,19] The observed J_c (2.3×10^6 A/cm^2) and average current density of 7.3×10^5 A/cm^2 at 2 K under a magnetic field of ≥3 T are slightly higher than that of the value of J_c (1.5×10^6 A/cm^2) and average J_c of 5.5×10^5 A/cm^2 at low temperature for Yb free $CeFeAsO_{0.9}F_{0.1}$ and other rare earth iron arsenides[20,21] indicating a strong flux pinning force, which arises due to Yb doping. The average critical current density of 7.3×10^5 A/cm^2 at low temperature (2 K) and in field of 3 to 5 T was calculated from the values obtained from field dependent J_c curves (Figure 4) (12.1, 10.32, 6.31, 4.98, and 3.01 A/cm^2).

For many recently reported ferropnictide superconductors, the thermally activated flux energy is one of the reasons for the broadening of resistivity peak. This may be due to a slow diffusion of magnetic vortices at certain temperature and magnetic field. To investigate the relation between thermal activation energy and field dependence resistivity, we use thermally activated flux flow model ($\rho = \rho_0 \exp[-U_0/k_B T]$), where U_0 is flux flow energy or pinning potential, ρ and ρ_0 are the normal state and residual resistivity, and K_B is the Boltzmann constant (1.380×10^{-23} m^2 kg s^{-2}). In Fig. 5, we have shown the plot of ln(ρ) vs T^{-1} (Arrhenius relation) and it could be clearly seen that U_0 is strongly combined with applied magnetic field. The value U_0 can be calculated from the slope

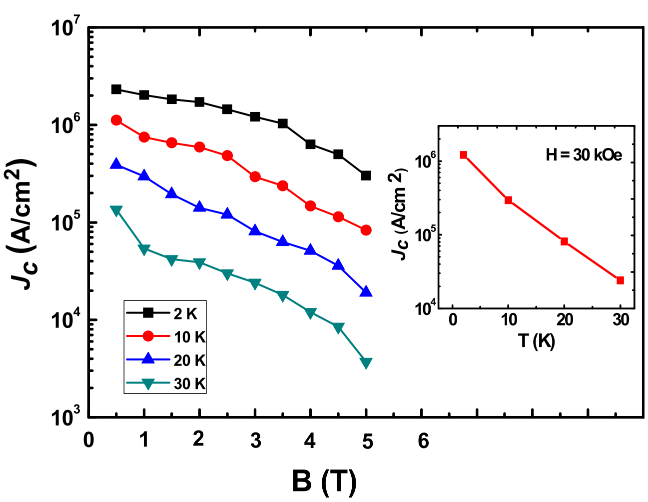

FIG. 4. Magnetic field dependence of critical current density (J_c) at different temperatures (2, 10, 20, and 30 K). Inset of figure denotes the variation of J_c with temperature at 30 kOe.

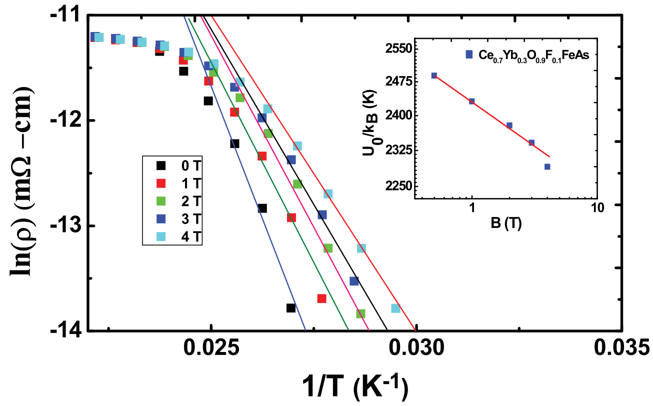

FIG. 5. Arrhenius plots of normal state of resistivity of $Ce_{0.7}Yb_{0.3}FeAsO_{0.9}F_{0.1}$. The activation energy (U_0) at an applied magnetic field is given by the slope from a linear fit. Inset shows magnetic field dependence of the activation energy (U_0).

of the line in Arrhenius plot. The magnetic field dependence of the activation energy (U_0) is shown in the inset of Fig. 5. From our experiment, the calculated activation energy (U_0/k_B) lies between ~2500 K and 2200 K in the magnetic field ranges of 0.5 T to 4 T. For $Ce_{0.7}Yb_{0.3}FeAsO_{0.9}F_{0.1}$, the obtained values of U_0 were seven times higher than that of $LaFeAsO_{0.85}F_{0.15}$ [Ref. 22] showing significant improvement of U_0 by Yb doping in place of Ce. This value of U_0 is three times larger than Bi2223 single crystal[23] at low magnetic field of 0.1 T. However, activation energy of $Ce_{0.7}Yb_{0.3}FeAsO_{0.9}F_{0.1}$ is low as compared to $BaFe_{1.9}Ni_{0.1}As_2$ and $Ba_{0.55}K_{0.45}Fe_2As_2$ (U_0/k_B = 5300 K and 9100 K at 0.1 T) single crystals.[24,25] Shahbazi *et al.* recently reported that a decrease in the activation energy (U_0/k_B) from 2000 K to 530 K at a field of 0.1 T was found in $CeFeAsO_{1-x}F_x$ (x = 0.1 and 0.2).[26] Inset of Fig. 5 shows gradual decrease in U_0 (~2500 K to 2200 K at 0.5 to 4 T) with increase in magnetic field for the Yb doped superconductor, which might be caused by strong vortex pinning forces co-existing with the applied magnetic field.

IV. CONCLUSIONS

We have discussed our studies on the upper critical field, critical current density, microstructure, and thermally activated flux energy of the $Ce_{0.7}Yb_{0.3}FeAsO_{0.9}F_{0.1}$ superconductor. Temperature dependent magnetoresistance studies show a significant improvement of upper critical field (H_{c2} (0) ~ 142 T) and high J_c value was found to be 2.3×10^6 A/cm^2 at 2 K under magnetic field of 0.5 T by using Bean's model. The estimated thermal activation energy of U_0/k_B ~ 2500 K at 0.5 T is slightly higher than those previously reported Sm, Nd, and Ce rare-earth iron pnictides. Our studies show that on substitution of smaller rare earth metal (Yb) in place of Ce in CeFeAsO/F successfully improves the critical current density and enhances magnetic flux pinning forces making this superconductor a potential candidate for superconducting applications.

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