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Enhancement in transition temperature and upper critical field of $\text{CeO}_{0.8}\text{F}_{0.2}\text{FeAs}$ by yttrium doping

J. Prakash,¹ S. J. Singh,² A. Banerjee,³ S. Patnaik,^{2,a)} and A. K. Ganguli^{1,a)}

¹Department of Chemistry, Indian Institute of Technology, New Delhi 110016, India

²School of Physical Sciences, Jawaharlal Nehru University, New Delhi 110067, India

³UGC-DAE Consortium for Scientific Research, University Campus, Khandwa Road, Indore 452017, India

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We report significant enhancement in superconducting properties of yttrium substituted $\text{Ce}_{1-x}\text{Y}_x\text{OFFeAs}$ superconductors. The polycrystalline samples were prepared by two step solid state reaction technique. X-ray diffraction confirmed tetragonal ZrCuSiAs structure with decrease in both a and c lattice parameters on increasing yttrium substitution (with fixed F content). With smaller ion Y in place of Ce, the transition temperature increased by 6 K. Yttrium doping also lead to higher critical fields as well as broader magnetization loops, particularly at elevated temperature. © 2009 American Institute of Physics. [doi:10.1063/1.3280046]

Shortly after the discovery of superconductivity in oxypnictides,¹ it was discerned that due to low transition temperature (T_c) and the deleterious influence of the grain boundaries, the possibility of technological application of these novel materials would be rather limited. The maximum $T_c \sim 55$ K thus far is reported in a samarium-based compound² but this is far below the liquid nitrogen benchmark. Moreover the pnictides seem to have all the negatives of high T_c cuprates particularly with respect to suppressed in-field intragranular pinning and weak link behavior across the grain boundaries.^{3,4} One favorable aspect of oxypnictides is the high upper critical field (H_{c2}) as a consequence of multiband effects.⁵ The important question of current relevance is the route to increase the transition temperature and critical current density in these materials without compromising on high upper critical field parameters. In this letter we report the successful synthesis of $\text{Ce}_{1-x}\text{Y}_x\text{O}_{0.8}\text{F}_{0.2}\text{FeAs}$ by simultaneous substitution of yttrium for cerium and fluorine for oxygen and study the superconducting properties in detail. We show that optimal Y addition can lead to significant increase in the transition temperature (T_c) and upper critical field (H_{c2}) as well as some signatures of improvement of the critical current density (J_c).

Polycrystalline samples with nominal compositions of $\text{Ce}_{1-x}\text{Y}_x\text{O}_{0.8}\text{F}_{0.2}\text{FeAs}$ and $\text{CeO}_{0.8}\text{F}_{0.2}\text{FeAs}$ were synthesized by a two step solid state method⁶ using high purity Ce, CeO_2 , Y_2O_3 , CeF_3 , and FeAs as starting materials. FeAs was obtained by reacting Fe chips and As powder at 800 °C for 24 h. The raw materials were taken according to stoichiometric ratio and then sealed in evacuated silica ampoules (10^{-4} torr) and heated at 900 °C for 30 h. The powder was then compacted (5 tons) and the disks were wrapped in Ta foil, sealed in evacuated silica ampoules and heated at 1100 °C for 30 h. All chemical manipulations were performed in a nitrogen-filled glove box. The samples were characterized by powder x-ray diffraction with $\text{Cu-K}\alpha$ radiation. Resistivity measurements were carried out using a Cryogenic 8 T Cryogen-free magnet in conjunction with a variable temperature insert. The inductive part of the mag-

netic susceptibility was measured using a tunnel diode based rf (2.3 MHz) penetration depth technique.⁷ Magnetization hysteresis loops and remanent magnetization of the samples were measured by a superconducting quantum interface device magnetometer.

Figure 1 shows the powder x-ray diffraction patterns for $\text{Ce}_{0.6}\text{Y}_{0.4}\text{O}_{0.8}\text{F}_{0.2}\text{FeAs}$ and $\text{CeO}_{0.8}\text{F}_{0.2}\text{FeAs}$. Majority of the observed reflections could be satisfactorily indexed on the basis of the tetragonal ZrCuSiAs type structure. Minor amount ($\sim 10\%$) of Y_2O_3 was observed as a secondary phase for Y-doped sample. The refined lattice parameters were found to be $a=3.9654(1)$ Å and $c=8.5803(3)$ Å for Y-doped sample and $a=3.988(3)$ Å and $c=8.607(8)$ Å for without Y-doped phase. The lattice parameters are smaller than the parent compound CeOFeAs [$a=3.996$ Å and $c=8.648$ Å (Ref. 8)] and the reduction in the lattice volume upon F and Y-doping indicates a successful chemical substitution. Both the lattice parameters for $\text{Ce}_{1-x}\text{Y}_x\text{OFFeAs}$ ($x=0.1, 0.2, 0.3,$ and 0.4) decrease with the increase in yttrium content but in this work we focus only on $x=0$ and 0.4 compositions. As shown in the inset of Fig. 1, the zero

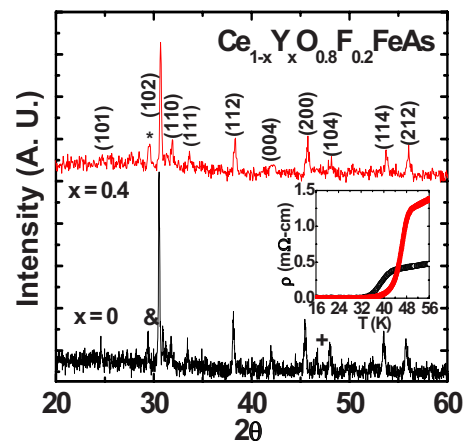


FIG. 1. (Color online) Powder x-ray diffraction patterns of (a) $\text{Ce}_{0.6}\text{Y}_{0.4}\text{O}_{0.8}\text{F}_{0.2}\text{FeAs}$ and (b) $\text{CeO}_{0.8}\text{F}_{0.2}\text{FeAs}$. The impurity phases are Fe_2As (+) and CeAs (&) for samples without Y and Y_2O_3 (*) for Y-doped sample. Inset of figure shows resistivity plot for $\text{CeO}_{0.8}\text{F}_{0.2}\text{FeAs}$ (○) and $\text{Ce}_{0.6}\text{Y}_{0.4}\text{O}_{0.8}\text{F}_{0.2}\text{FeAs}$ (●).

^{a)}Authors to whom correspondence should be addressed. Electronic addresses: spatnaik@mail.jnu.ac.in and ashok@chemistry.iitd.ernet.in.

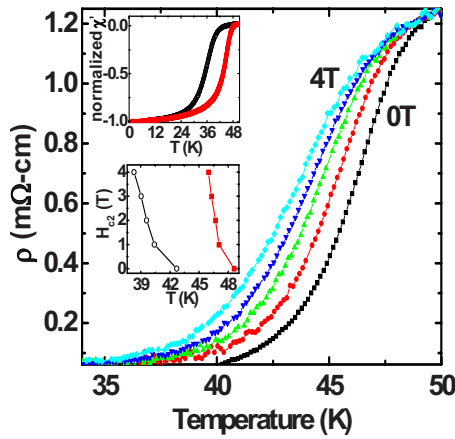


FIG. 2. (Color online) Temperature dependence of the electrical resistivity of $\text{Ce}_{0.6}\text{Y}_{0.4}\text{O}_{0.8}\text{F}_{0.2}\text{FeAs}$ under varying magnetic fields (0–4 T). Upper inset shows the inductive part of susceptibility as a function of temperature and the lower inset depicts the temperature dependence of upper critical field (H_{c2}) as a function of temperature for $\text{Ce}_{0.6}\text{Y}_{0.4}\text{O}_{0.8}\text{F}_{0.2}\text{FeAs}$ (■) and $\text{CeO}_{0.8}\text{F}_{0.2}\text{FeAs}$ (○).

field onset transition temperature for $x=0.4$ is estimated to be 48.6 K as compared to $x=0$ case where it is 42.7 K.⁹ Surprisingly for the Ce(O/F)FeAs, the T_c is suppressed on increasing external pressure and is lowered to 1.1 K at 265 kbar.¹⁰ On the contrary we find that the T_c of Ce(O/F)FeAs enhances on increasing the chemical pressure by substituting smaller Y^{3+} ion in place of Ce^{3+} . The onset of diamagnetic behavior below T_c is confirmed from the inductive part of rf susceptibility as shown in the upper inset of Fig. 2. The onset transition temperature (T_c) determined is 47.5 and 40.5 K for $\text{Ce}_{0.6}\text{Y}_{0.4}\text{O}_{0.8}\text{F}_{0.2}\text{FeAs}$ and $\text{CeO}_{0.8}\text{F}_{0.2}\text{FeAs}$, respectively. This bulk magnetization shows a slightly lower T_c as compared to the resistivity data. The residual resistivity ratio (RRR = ρ_{300}/ρ_{50}) is estimated to be 6.6 and 3.5 for $\text{CeO}_{0.8}\text{F}_{0.2}\text{FeAs}$ and $\text{Ce}_{0.6}\text{Y}_{0.4}\text{O}_{0.8}\text{F}_{0.2}\text{FeAs}$, respectively. We emphasize that the onset of superconductivity in $\text{Ce}_{0.6}\text{Y}_{0.4}\text{O}_{0.8}\text{F}_{0.2}\text{FeAs}$ represents the highest transition temperature in Ce-based oxypnictides synthesized at ambient pressure.

The temperature dependence of the resistivity under varying magnetic fields is shown in Fig. 2 for sample $\text{Ce}_{0.6}\text{Y}_{0.4}\text{O}_{0.8}\text{F}_{0.2}\text{FeAs}$. Using a criterion of 90% of normal state resistivity, the estimated H_{c2} - T phase diagram for both samples is shown in lower inset of Fig. 2. The Werthamer Helfand Hohenberg formula,¹¹ gives $H_{c2}(0) = -0.693 T_c (dH_{c2}/dT)_{T=T_c}$. The slope of dH_{c2}/dT estimated from the H_{c2} - T plots is -2.67 and -1.45 T/K for Y-doped and without Y-doped samples, respectively. Correspondingly, we find $H_{c2}(0) \sim 90$ T and 43 T for $\text{Ce}_{0.6}\text{Y}_{0.4}\text{O}_{0.8}\text{F}_{0.2}\text{FeAs}$ and $\text{CeO}_{0.8}\text{F}_{0.2}\text{FeAs}$ respectively. Evidently, we are able to significantly increase upper critical field by simultaneous doping of yttrium and fluorine.¹²

The inset in Fig. 3 shows a characteristic magnetization loop for the sample $\text{Ce}_{0.6}\text{Y}_{0.4}\text{O}_{0.8}\text{F}_{0.2}\text{FeAs}$. Both samples showed asymmetric $M(H)$ loop similar to polycrystalline La iron oxypnictides.³ The loop width [$\Delta m(H)$] is small, which is a general feature of oxypnictides and could be due to either poor intergrain connectivity or weak intragrain pinning.³ The magnetic field dependence of Δm at 4.2, 15, and 30 K for both samples are compared in Fig. 3. The samples were of different shapes and we have normalized Δm by the weight of the specimen (14.8 mg for CeOFFeAs

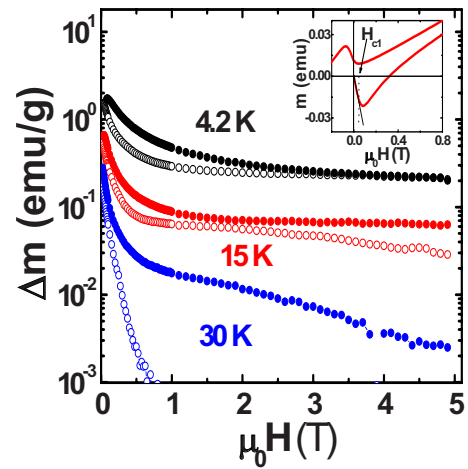


FIG. 3. (Color online) Magnetic field dependence of magnetization hysteresis loop width (Δm) at 4.2, 15, and 30 K for $\text{Ce}_{0.6}\text{Y}_{0.4}\text{O}_{0.8}\text{F}_{0.2}\text{FeAs}$ (closed symbols) in comparison with $\text{CeO}_{0.8}\text{F}_{0.2}\text{FeAs}$ (open symbols). Inset shows the magnetization hysteresis loop at 4.2 K for polycrystalline $\text{Ce}_{0.6}\text{Y}_{0.4}\text{O}_{0.8}\text{F}_{0.2}\text{FeAs}$ and defines procedure for determination of H_{c1} .

specimen and 18 mg for CeYOFFeAs). A clear improvement in magnetization loop width is observed for CeYOFFeAs particularly at elevated temperature.

To analyze relative contribution from inter and intragrain regions to $\Delta m(H)$ and consequently to the corresponding critical state current density, in Fig. 4 we show the remanent moment as a function of increasing applied field. The samples were studied (at $T=5$ K) under cycles of increasing magnetic field followed by removal of the field and measurement of remanent moment.^{3,13} When external field is applied, first the flux enters into the intergranular region followed by grain penetration above lower critical field H_{c1} . The remanent moment equals the product of local current density multiplied by current loop size and therefore a single step variation m_R versus H curve indicates predominant role of intragrain supercurrents. We also observe that the remanent moment deviates from zero at ~ 7 and 20 mT for $\text{CeO}_{0.8}\text{F}_{0.2}\text{FeAs}$ and $\text{Ce}_{0.6}\text{Y}_{0.4}\text{O}_{0.8}\text{F}_{0.2}\text{FeAs}$, respectively. This implies some improvement in intergrain pinning as well in the Y doped sample. In the inset of Fig. 4, we plot $H_{c1}(T)$ derived from the departure from linearity in low field M - H loop (elucidated in the inset of Fig. 3). The technique is

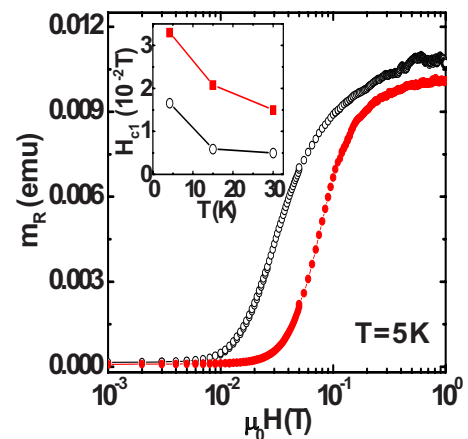


FIG. 4. (Color online) The field-dependent remanent magnetization at 5 K for both samples. The inset shows the temperature dependence of lower critical field (H_{c1}).

strictly applicable in slab geometry¹⁴ and typically overestimates H_{c1} , but there is definite relative increase in the lower critical field for the Y doped sample.

In summary, we have demonstrated substantial enhancement in transition temperature, critical fields, and magnetization loop widths by simultaneous doping of Y in place of Ce and F in place of O in semimetal CeOFeAs. This multitudinous beneficial effect is caused by three independent parameters; higher chemical pressure, selective tuning of interband scattering, and superior pinning properties of Y_2O_3 .

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