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The effect of partial Lu doping on magnetic behaviour of YBCO (123) superconductors

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ABSTRACT

We have investigated the effect of Lutetium doping on YBCO prepared by conventional solid-state reaction method by means of XRD, AC susceptibility and DC magnetization measurements and theoretical analysis. AC susceptibility measurements for sintered YBCO pellets have been performed as a function of temperatures at constant frequency and AC field amplitude in the absence of DC bias field. The critical current densities as a function of temperature have been estimated from the AC susceptibility data. DC magnetization measurements were done at 20 K and 77 K upon zero field cooling (ZFC) process. 50% Lu substitution for Y on YBCO superconductors improves the critical current density and lower critical field H_{c1} at low temperature region (at 20 K).

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1. Introduction

Since the discovery of Y–Ba–Cu–O compound system, it has been widely studied for improving the superconducting properties by many researchers. A great number of papers have been published on adding elements in Y123 system for increasing the critical temperature (T_c) and critical current density (J_c) values of YBCO samples. The main factors reducing the J_c in HTSC are grain boundaries and poor flux pinning [1]. The low values of the grain-boundary critical current densities J_c in polycrystalline samples are a significant problem for large-current applications, for which the enhancements of J_c is a key issue [2]. Some authors proposed to solve this problem using a three-fold approach: Through (a) grain alignment [3], (b) grain-boundary doping [4], and (c) optimization of the microstructure to maximize the effective grain-boundary area [5]. It was found that Y can be replaced with various rare earth elements (Re) [6–21], including Lu [6–9,21] without any significant change in T_c for Re–Ba–Cu–O (123) system (Re = La, Nd, Sm, Eu, Gd, Dy, Ho, Er, Tm, Yb and Lu, from the largest to the smallest radius). Re elements substitute for both Y and Ba sites in Y123 compounds. In the case of trivalent Re ions substitution for Y^{3+} and Ba^{2+}, there is a strong correlation between the solution energies and the size of dopant, the solution energy increases with increasing ionic radius. The Re ions towards the Lu end of the lanthanide series are comparable in size to Y^{3+} and a degree of mixing among the yttrium and barium sites might be expected to occur [22]. Moreover, Lu cannot wholly substitute for Y [7,8], probably its small radius but it can be incorporated along with co-substitution of a small amount of Sr for Ba [23].

Raychaudhuri et al. [9] reported that partial Lu substituted YBCO ($Y_{0.75}Lu_{0.25}Ba_2Cu_3O_7$) sample shows much more Meissner effect than pure YBCO measured by DC magnetic susceptibility data at 20 K. It was also reported that after the further annealing treatment, the sample doped with 0.5 wt% Lu oxide, prepared by top-seeding-melt-texture-growth process, exhibit better critical current density than the undoped Y123 sample [21].

In this work, we present the results of XRD, AC susceptibility and DC magnetization measurements on pure and partial Lu doped YBCO superconducting samples and investigate the effect of Lu doping on the superconducting parameters such as lattice constants, transition temperature, and critical current density etc., in the samples studied.

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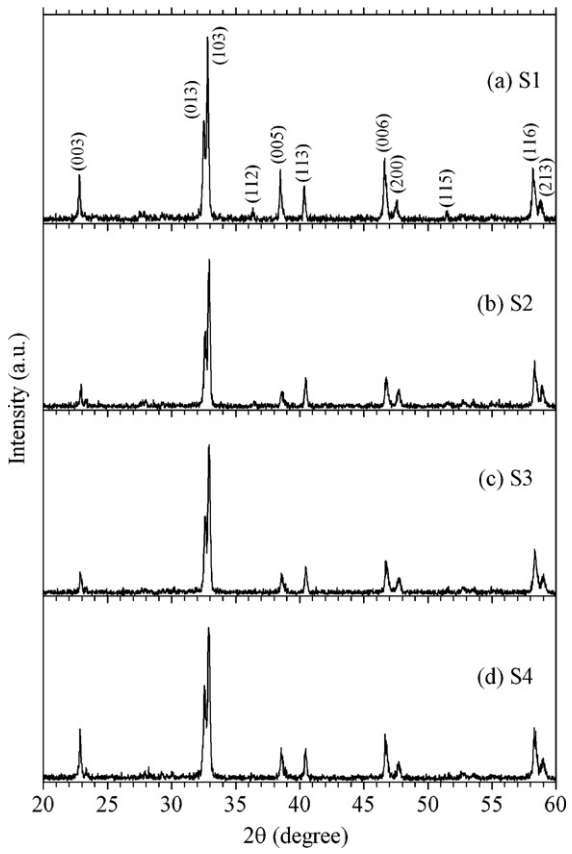

Fig. 1. XRD patterns of samples prepared by the conventional solid-state reaction method: (a) S1, (b) S2, (c) S3 and (d) S4 samples. The indexed peaks are those of YBCO.

2. Experimental

The samples with nominal composition $Y_{1-x}Lu_xBa_2Cu_3O_y$ (where $x = 0$, 0.2, 0.4, 0.5) were prepared by the conventional solid-state reaction method. We use the sample labels S1, S2, S3, and S4 to refer the samples with $x = 0$; 0.2; 0.4; 0.5, respectively. Powders of Y_2O_3, $BaCO_3$, CuO and Lu_2O_3 were thoroughly mixed in the appropriate amounts and calcined at 890 °C for 24 h. After calcination, the powders were mixed for 1 h and then sintered at 900 °C for 24 h. The powders were reground and pressed into pellets of 13 mm diameter at 375 MPa. The pellets were resintered at 900 °C for 12 h and cooled to 450 °C at 2 °C/min. The pellets were maintained at this temperature for 2 h with oxygen flowing. Then the pellets were cooled to room temperature at a cooling rate of 1 °C/min by flowing oxygen at temperatures between 450 and 250 °C.

The X-ray diffraction (XRD) data were taken using Rigaku D/Max-IIIC diffractometer with Cu Kα radiation over the range of 20–60° with a scan speed of 0.2°/min at room temperature. The orthorhombic lattice parameters (a, b and c) were calculated from (0 0 3), (0 1 3), (1 0 3), (0 0 5), (0 0 6), (2 0 0), (1 1 6) and (2 1 3) peaks using least square methods.

The AC susceptibilities and DC magnetization measurements were carried out with a commercial Quantum Design Physical Property Measurement System (PPMS) Model 6000, ACMS and VSM option respectively. The dimensions ($2t \times 2w \times l$) of the samples S1, S2, S3, and S4 are 2.2 mm × 2.35 mm × 2.5 mm, 2.29 mm × 2.32 mm × 2.10 mm, 2.36 mm × 2.37 mm × 2.08 mm and 2.05 mm × 2.23 mm × 2.43 mm, respectively. The sample was mounted with its length along the direction of the collinear magnetic fields. The signal due to the in-phase and out-of-phase magnetization was taken simultaneously. DC magnetization measurements were carried out at 20 K and 77 K upon zero field cooling (ZFC) process.

3. Results and discussion

Fig. 1 shows the X-ray diffraction patterns of the samples (a) S1, (b) S2, (c) S3 and (d) S4. It can be seen from the XRD data that the samples have a single phase of orthorhombic Y123 structure. There are hardly any impurities in all the samples. Because the solubility limits of Lu in YBCO is $x = 0.5$ maximum and small content of second phases appears when $x > 0.5$, we have focused only $x = 0$–0.5 substitution range. There is very little change in lattice parameters. However, the orthorhombic strain $\Delta = (b - a)/(b + a)$ increases with the Lu substitution, where a and b are the lattice parameters of the samples. The lattice parameters, orthorhombic strains and transition temperatures for the samples are listed in Table 1. It can be seen from Table 1 that the lattice parameters of the Lu doped samples (S2, S3, and S4) are smaller than those for S1 (pure YBCO). This can be attributed to effect of Lu substitution for Y since the ionic radius of Lu (0.977 Å) is smaller than the ionic radius of Y (1.019 Å). Raychaudhuri et al. [9] also reported small unit-cell parameters of partial Lu substituted YBCO ($Y_{0.75}Lu_{0.25}Ba_2Cu_3O_7$) system.

Table 1
Lattice parameters, orthorhombicity and T_c for the samples.

Sample	S1	S2	S3	S4
a (Å)	3.822	3.812	3.808	3.809
b (Å)	3.882	3.875	3.874	3.877
c (Å)	11.676	11.648	11.645	11.657
$\Delta = (b-a)/(b+a)$	8.007×10^{-3}	8.195×10^{-3}	8.591×10^{-3}	8.950×10^{-3}
T_c (K)	92	91.5	90	89.5

We display the temperature behaviour of the ac susceptibility for the samples at 480 A/m ac field amplitude (rms) in Fig. 2, where $H_b = 0$ (H_b: DC field). Fig. 2(a) displays the curves χ' versus T and 1b) χ'' versus T at $f = 1$ kHz. We normalized experimental ac susceptibility data $\chi''(T)$ and $\chi'(T)$ to the $|\chi'|$ at the lowest temperature and the lowest field amplitude for each sample since the demagnetizing correction would cause $\chi' = -1$ for low enough temperature at low field amplitude. The diamagnetic transition temperatures are

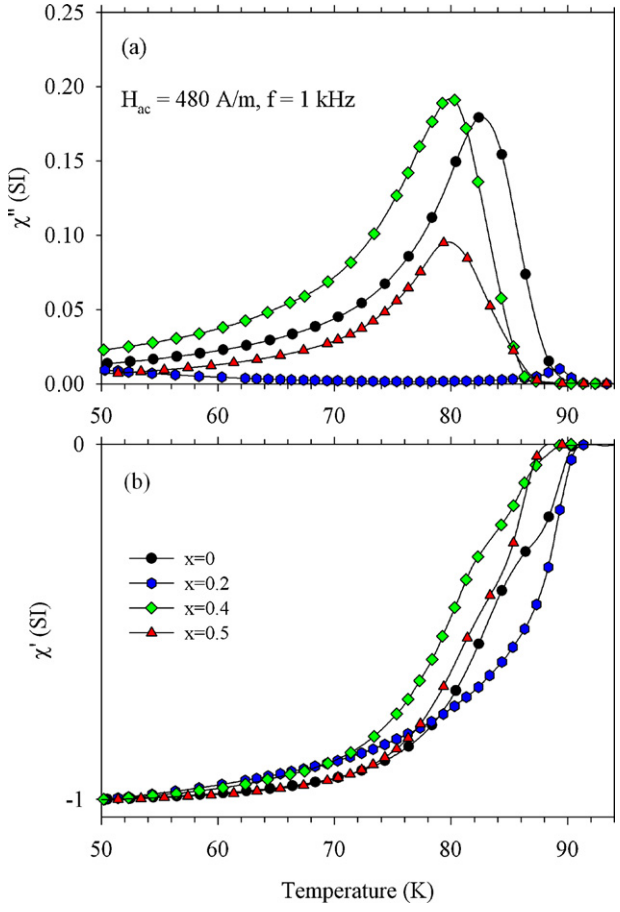

Fig. 2. Plot of (a) χ'' versus temperature and (b) χ' versus temperature at $f = 1$ kHz and $H_{ac} = 480$ A/m for the samples studied.

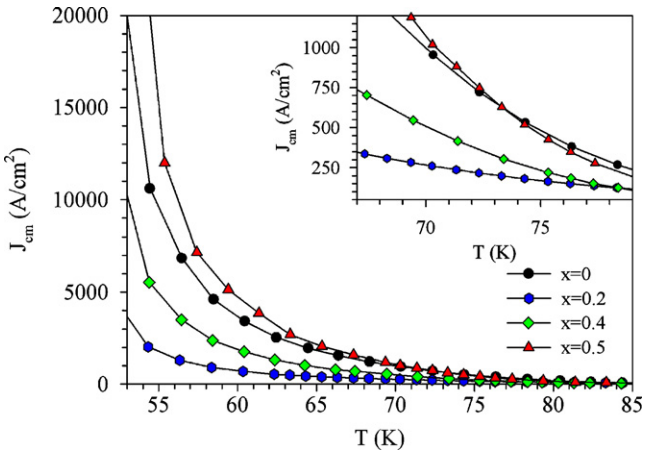

Fig. 3. Intergranular critical current density J_{cm} of the samples, as a function of temperature at H_{ac} = 480 A/m (rms) and f = 1 kHz. The inset shows the temperature dependence of J_{cm} in different temperature range.

found to be 92 K, 91.5 K, 90 K and 89.5 K for the samples S1, S2, S3, and S4 respectively. The transition temperature T_c decreases monotonically with Lu concentration x. The substitution of Lu^{3+} ions for Y^{3+} ions possibly introduces difference in the electronic state in the CuO_2 plane and affects the transition temperature T_c. Hor et al. [7] reported the transition temperature of the $LuBa_2Cu_3O_{6+x}$ structure was 85 K measured by resistive method. Tarascon et al. [8] reported the transition temperature of the $LuBa_2Cu_3O_{7-x}$ system was 88.2 K by using resistive method.

Fig. 3 shows the temperature dependence of intergranular critical current density J_{cm}. For these values of J_{cm}, we have used AC susceptibility data taken at H_{ac} = 480 A/m (rms), f = 1 kHz. In hysteretic type II superconductors, when the flux lines fully penetrate the material, i.e., when H_a ($H_a = \sqrt{2}H_{ac}$ (rms)) is equal to the first full penetration field H_*, the losses reach a maximum [24] for infinite cylinder geometry. It is noted that residual trapped flux calculations [25] reveal that the results for the square sample are identical to that of the infinite cylinder except that the factor of 4 should be replaced by π. According to Bean model [26], the critical current density at the peak temperature T_p, can be written $J_c(T_p) = H_*/a \approx H_*/\sqrt{ab}$, and where the cross-section of the rectangular bar-shaped sample is $2a \times 2b$. Under these approximations and using Clem's following equations [24], we have estimated and displayed the temperature dependence of intergranular critical current density J_{cm} for our samples in Fig. 3:

$$\chi'(T) = -1 + \frac{J_c(T_p)}{J_c(T)}\left(1 - \frac{5}{16}\frac{J_c(T_p)}{J_c(T)}\right) \quad \text{for } T < T_p \tag{1}$$

$$\chi'(T) = \frac{-5}{16}\frac{J_c(T)}{J_c(T_p)} \quad \text{for } T > T_p \tag{2}$$

We note that sample S4 has larger J_{cm} values than other samples at low temperatures. Appropriate doping provides an improvement in the transport properties of grain boundaries in high-T_c superconductors [27]. To explore the possibility of increasing J_c, some workers investigated and reported on the mechanisms controlling the grain-boundary properties [28–30]. Their work proves that the established upper limit to the grain-boundary critical current density for a given misorientation angle between the grains can be overcome and further increases in J_c will be realized by optimizing the dopant, its concentration, and the deposition conditions. We also believe that Lu doping may have changed the microstructure of the boundary by changing the grain size and grain-boundary area, which will have an influence on J_c.

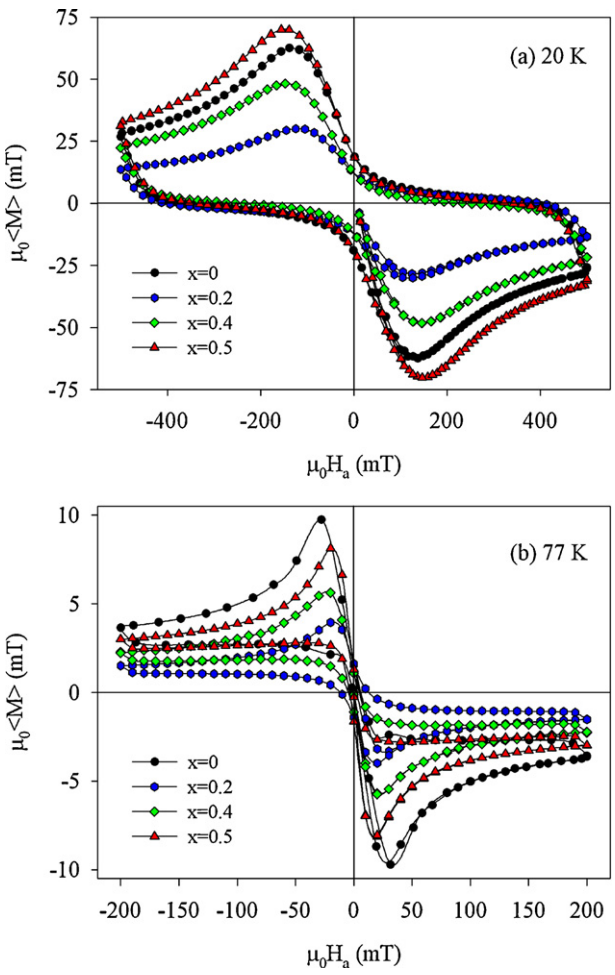

Fig. 4. Magnetization hysteresis curves for our YBCO samples at (a) 20 K and (b) 77 K.

The inset in Fig. 3 displays the high temperature range of J_{cm} in enlarged scale. The inset shows that below 73 K, sample S4 has higher J_{cm} values than samples S1, S2 and S3, but above 73 K, sample S1 has higher J_{cm} values. Schmehl et al. [31] reported that partial doping of the Y^{3+} in Y123 with Ca^{2+} increases grain-boundary J_c values substantially, but only at temperatures much lower than 77 K. As their experiments have proven, appropriate doping enhances J_c at low temperatures, for temperatures close to T_c, the doping-induced reduction of T_c lowers J_c. Hammerl et al. [4] found similar behaviour in partial (x = 0.3) Ca doped Y123 system. Their Ca doped samples showed a very large critical current density than undoped sample only at temperatures lower than ~60 K. This behaviour can be also explained theoretically by temperature dependence of the critical current density given by the power law relation $J_{cm} = J_{cm0}(1 - T/T_c)^s$. For pure YBCO and Lu doped YBCO samples, different J_{cm0}, T_c and s exponent values may cause this behaviour. The DC magnetization measurements of the samples at 20 K and 77 K also support this result.

In Fig. 4, we present the magnetization curves for the samples at (a) 20 K and (b) 77 K with the applied field, $\mu_0 H_a$, increasing from zero to a maximum value of 0.5 T (0.2 T for 77 K), then decreasing to zero, reversing in sign to a maximum negative value of 0.5 T (0.2 T for 77 K), and finally increasing again to 0.5 T (0.2 T for 77 K). Each curve is called the envelope of the major hysteresis loop. It is well established that the width and behaviour of this hysteresis loop is the manifestation of the bulk critical current density in type II superconductors and its dependence on the magnetic flux density.

The wider hysteresis loop is, the higher bulk critical current density will be. Hence it can be readily seen from Fig. 4 that sample S4 has the highest bulk critical current density at 20 K. However M-H loop at 77 K indicate that Lu doping to YBCO caused to decrease the bulk critical current density. This conclusion is consistent with the one obtained from AC susceptibility result and attributed to having different temperature exponent s of the critical current density, which could be related to appropriate doping and mechanisms controlling grain-boundary properties [27–31]. It is also well known that in some type II superconductors there exists Meissner currents circulating along the periphery of the sample, which shifts the hysteresis loop to diamagnetic quadrant [32]. In our samples under study, we have seen this effect indicating that there exists Meissner current in addition to bulk critical current. Using the descending and ascending branch of the hysteresis loop, we estimated Meissner currents per unit height of the samples studied. It was found that 50% Lu doping to YBCO increased the Meissner current and lower critical field in the low temperature region as reported in Ref. [9]. The detailed theoretical analysis including the currents associated with the surface barrier will be published elsewhere.

4. Conclusions

We have measured AC susceptibility and the DC magnetization and of our pure YBCO and Lu doped YBCO samples prepared by the conventional solid-state reaction method. AC susceptibility measurements have been performed at constant frequency and AC field amplitude. Employing Bean's and Clem's model, we determined the temperature dependent critical current density. We have measured the DC magnetization on the pure YBCO and Lu doped YBCO samples at 20 K and 77 K by a VSM. We have found that 50% Lu doping to YBCO increased the bulk critical current density and lower critical field H_{c1} at 20 K and decreased the critical temperature T_c.

Acknowledgements

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