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The Study of Magnetic and Transport Properties in Normal State $Eu_{1.85+y}Ce_{0.15-y}Cu_{1-y}Fe_yO_{4+\alpha-\delta}$

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Abstract**—**The effect of partially substitution of magnetic impurity Fe for Cu to the magnetic and transport properties in electron-doped superconducting cuprates of $Eu_{1.85+y}Ce_{0.15-y}Cu_{1-y}Fe_yO_{4+\alpha-\delta}$ (ECCFO) with y = 0, 0.010, 0.020, and 0.050 has been studied, in order to investigate the mechanism of magnetic and transport properties of ECCFO in normal-state. Magnetic properties are investigated by DC magnetic-susceptibility measurements that carried out at low temperatures down to 2 K using a standard SQUID magnetometer in a magnetic field of 5 Oe on field cooling. Transport properties addressed to electron mobility, are extracted from radius of electron localization calculated from temperature dependence of resistivity. For y = 0, temperature dependence of dc magnetic-susceptibility (χ) indicated the change of magnetic behavior from paramagnetic to diamagnetic below 15 K. Above 15 K, all samples show paramagnetic behavior with the values of magnetic moment in every volume unit increased with increasing y. Electron mobility decreased with increasing y.

Keywords**—**DC magnetic-susceptibility, electron mobility, $Eu_{1.85+y}Ce_{0.15-y}Cu_{1-y}Fe_yO_{4+\alpha-\delta}$, normal state.

I. Introduction

The first material of high-T_c superconductors (HTSC) discovered in 1986 was single-layer cuprates called 214 cuprates. These 214 cuprates consist of two doping systems, namely, hole-doped and electron-doped superconductor. The electron-hole doping symmetry in HTSC has been one of great interests in relation to the mechanism of superconductivity. The studies of physical properties through the partially substitution of impurities for Cu in both systems are one of important ways to elucidate the electron-hole doping symmetry [1]–[4]. In the electron-doped system, the superconductivity is suppressed through the substitution of magnetic impurity Ni for Cu more markedly than through the nonmagnetic Zn substitution [5]. In hole-doped system, on the other hand, the substitution of nonmagnetic Zn for Cu suppressed superconductivity more markedly than magnetic impurity Ni substitution [1], [6]. Using another substitution of magnetic impurity Fe for Cu in hole-doped system, it has been found that the magnetic transition temperature and magnetic correlation are enhanced through the 1% Fe substitution in a wide range of hole concentration where superconductivity appears in Fe-free $La_{2-x}Sr_xCuO_4$ [7]. To our knowledge, however, complete report of the effects of substitution of magnetic impurity Fe for Cu to the magnetic properties in the electron-doped cuprates has not yet been available, so that clear conclusion for comparing the effect of impurity in electron- and hole-doped system has not yet been obtained.

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In this paper, we investigate effects of partially substitution of magnetic impurity Fe for Cu to the magnetic and transport properties in electron-doped superconducting cuprates of $Eu_{1.85+y}Ce_{0.15-y}Cu_{1-y}Fe_yO_{4+\alpha-\delta}$ (ECCFO).

II. Experimental

A. Sample Preparation

Polycrystalline samples of $Eu_{1.85+y}Ce_{0.15-y}Cu_{1-y}Fe_yO_{4+\alpha-\delta}$ with y = 0, 0.010, 0.020, and 0.050 were prepared by ordinary solid-state reaction method with some details procedure are described in our previous reports [8]. With increasing the concentration of Fe, we decreased the concentration of Ce in order to keep the charge carrier in the optimum-doped condition of 0.15. As-grown samples of ECCFO were checked by powder x-ray diffraction to be single phase and post-annealed in flowing Ar gas of high purity (6N) at various temperatures from 930 to 850 ^oC for 8 – 20 h, in order to remove the excess oxygen (α) at the apical site. The reduced oxygen content (δ) was estimated from the weight change before and after annealing. The values of δ are varying between 0.00896 and 0.462.

B. Characterization

DC magnetic-susceptibility (χ) measurements were carried out at low temperatures down to 2 K using a standard SQUID magnetometer (Quantum Design, Model MPMS-XL5) in a magnetic field of 5 Oe on field cooling.

The resistivity was measured with DC current in four-probe configuration from 4.2 tp 300 K.

III. Results and Discussion

Superconducting samples in impurity-free electron-doped superconducting cuprates are obtained when the value of δ is nearly equal to the value of α. However, we cannot control the value of δ well. For the present study, we obtained a set of impurity-substituted samples with δ values from 0.01 to 0.14. From our previous experiments, the best δ values getting high T_c superconductors are in the range of 0.02 to 0.09.

Fig. 1 shows Temperature dependence of dc magnetic-susceptibility (χ) on field cooling at 5 Oe for $Eu_{1.85+y}Ce_{0.15-y}Cu_{1-y}Fe_yO_{4+\alpha-\delta}$ with y = 0 - 0.030 and δ values from 0.0252 to 0.0665. For impurity-free samples of y = 0, diamagnetic behavior with χ smaller than 0 is observed starting from about 15 K. This temperature can be defined as T_c onset for this sample. For y = 0.005, T_c onset decreases to be around 10 K. The trace of superconductivity disappeared at y ≥ 0.020.

The volume fraction of the superconducting state, V_{SC}, has been estimated from the absolute value of χ at 2 K. We choose the value of χ at 2 K of impurity-free samples as corresponding to 100 % of V_{SC}. Then, V_{SC} for each impurity-substituted samples is estimated as each value of χ at 2 K divided by that of the impurity-free sample. V_{SC} decreases markedly from 100 % to below 5 % with only 1 % impurity substitution. V_{SC} is completely zero when the concentration of Fe is larger than 2 %. From these results, it is found that partially substitution of magnetic impurity Fe for Cu effectively decreased superconducting properties of electron-doped superconductor $Eu_{2-x+y}Ce_{x-y}Cu_{1-y}Fe_yO_{4+\alpha-\delta}$.

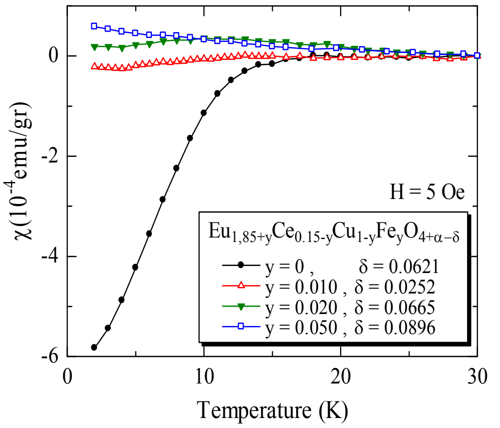

Fig. 1 Temperature dependence of dc magnetic-susceptibility (χ) on field cooling at 5 Oe for $Eu_{1.85+y}Ce_{0.15-y}Cu_{1-y}Fe_yO_{4+\alpha-\delta}$ with y = 0, 0.010, 0.020, 0.050 and δ values from 0.0252 to 0.0896

Above 15 K, all data show paramagnetic-like behavior. The dc magnetic-susceptibility (χ) dependent of temperature (T) is defined as shown in (1):

$$\chi = \frac{C}{T}, \quad (1)$$

where C is a Curie constant. The susceptibility (χ) also can be expressed as shown in (2):

$$\chi = \frac{\mu_0 \cdot n \cdot m^2}{3 \cdot k_B \cdot T} \quad (2)$$

where m is the magnetic moment per atom, n is the number of atoms per unit volume, k_B is Boltzmann's constant. So that, the Curie constant can be expressed as shown in (3) and the magnetic moment per atom per unit volume can be expressed as shown in (4):

$$C = \frac{\mu_0 \cdot n \cdot m^2}{3 \cdot k_B} \quad (3)$$

$$n \cdot m^2 = \frac{3 \cdot k_B \cdot C}{\mu_0} \quad (4)$$

Fig. 2 shows dc magnetic susceptibility (χ) versus 1/T for $Eu_{2-x+y}Ce_{x-y}Cu_{1-y}Fe_yO_{4+\alpha-\delta}$ with y = 0, 0.010, 0.020, 0.050 and δ values from 0.0252 to 0.0896 in the range temperature between 15 K and 30 K. Linear combination is well fitted for all data. The gradient value of all data can be extracted to find the value of Curie Constant (C) and magnetic moment per atom per unit volume.

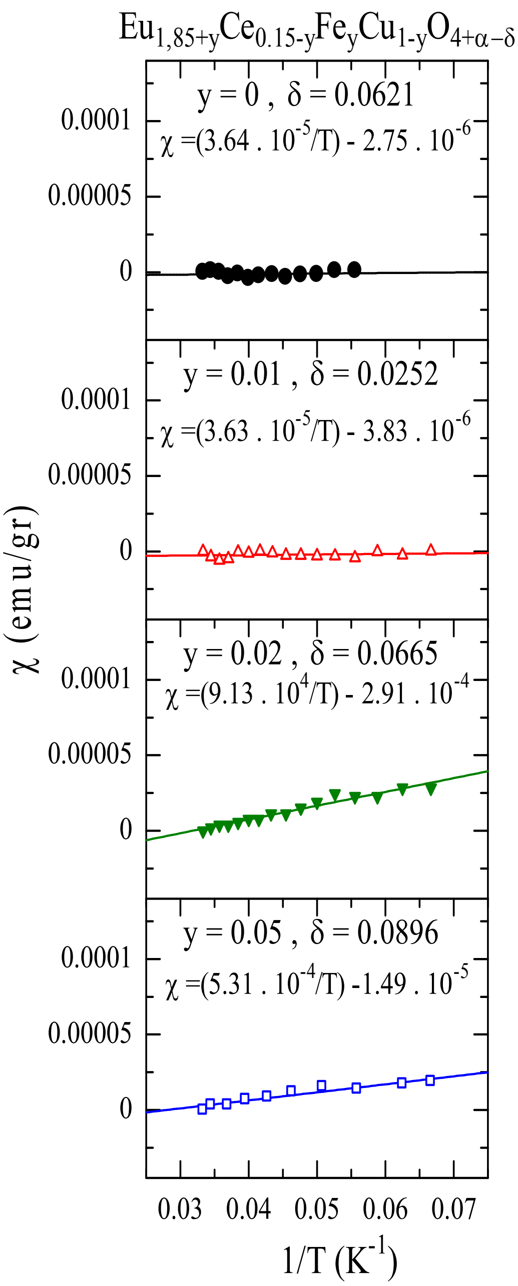

Fig. 2 Magnetic-susceptibility (χ) versus 1/T for $Eu_{1.85+y}Ce_{0.15-y}Cu_{1-y}Fe_yO_{4+\alpha-\delta}$ with y = 0, 0.010, 0.020, 0.050 and δ values from 0.0252 to 0.0896

Table I shows the value of Curie Constant (C) and magnetic moment per atom per unit volume ($n \cdot m^2$). There is no significant changing of the value of C and $n \cdot m^2$ when y = 0.01 is prepared in the samples. However, the value of C and $n \cdot m^2$ increase one order magnitude when y = 0.02 and 0.05 are applied. It is probably due to the contribution of orbital moment of high spin of Fe to all magnetic moments in the samples.

TABLE I
THE VALUE OF CURIE CONSTANT (C), AND MAGNETIC MOMENT/UNIT VOLUME

y	C	$n \cdot m^2$
0	$3.64 \cdot 10^{-5}$	$1.20 \cdot 10^{-21}$
0.010	$3.63 \cdot 10^{-5}$	$1.20 \cdot 10^{-21}$
0.020	$9.13 \cdot 10^{-4}$	$3.01 \cdot 10^{-20}$
0.050	$5.31 \cdot 10^{-4}$	$1.75 \cdot 10^{-20}$

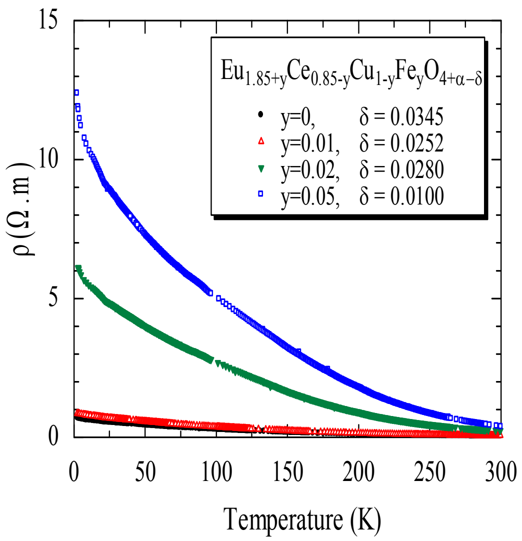

Fig. 3 Temperature dependence of resistivity ρ for $Eu_{1.85+y}Ce_{0.15-y}Cu_{1-y}Fe_yO_{4+\alpha-\delta}$ with y = 0, 0.010, 0.020, 0.050 in various δ values

Fig. 3 shows temperature dependence of electrical resistivity ρ for $Eu_{2-x+y}Ce_{x-y}Cu_{1-y}Fe_yO_{4+\alpha-\delta}$ with y = 0, 0.010, 0.020, 0.050 and various δ values. There is no trace of superconductivity observed in all data even for y=0. We suggested that it is because of surface problem in our samples since we performed our measurement in bulk system. Another possible reason for absence of the trace of superconductivity is because the excess oxygen in the samples is not removed well. In dc susceptibility data, the trace of superconductivity is observed when the value of δ is larger than 0.0621 for y = 0.

It is found that increasing of Fe concentration will increase the value of ρ. To analyze Fig. 3, we applied variable range hopping mechanism as express in (5) [9]:

$$\rho(T) = \rho_0 \exp[\left(\frac{T_0}{T}\right)^{\frac{1}{4}}], \quad (5)$$

where ρ_0 and T_0 are resistivity and temperature characteristic in the ground state. T_0 can be equal to $\frac{1}{r^3}$ and $\frac{1}{r^3}$ is equal to $\frac{1}{\mu}$ as described in (6) and (7), respectively:

$$T_0 \sim \frac{1}{r^3}, \quad (6)$$

$$\frac{1}{\mu} \sim \exp\left(\frac{1}{r^3}\right)^{\frac{1}{4}}, \quad (7)$$

where r is radius of localization of electron, and μ is electron mobility.

Fig. 4 shows $(1/T)^{1/4}$ versus Ln ρ for $Eu_{2-x+y}Ce_{x-y}Cu_{1-y}Fe_yO_{4+\alpha-\delta}$ with y = 0, 0.010, 0.020, 0.050 and various δ values at temperature range between 15 K to 30 K. It is found that Ln ρ is proporsional to $(1/T)^{1/4}$. The gradient of all data can describe radius of localization and mobility of electron.

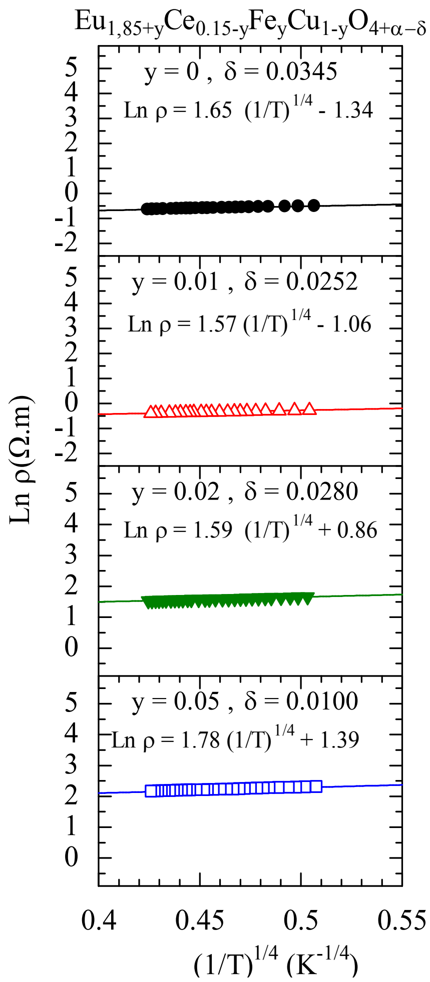

Fig. 4 The dependence of (1/T)1/4 versus Ln ρ for $Eu_{2-x+y}Ce_{x-y}Cu_{1-y}Fe_yO_{4+\alpha-\delta}$ with y = 0, 0.010, 0.020, 0.050 and various δ values at temperature range between 15 K to 30 K

Fig. 5 shows the y value of concentration dependence of (a) radius of localization of electron (r), and (b) electron mobility (μ). Both r and μ are slightly increase for y = 0.01. With increasing y, r and μ decrease. Below y = 0.01, the electron mobility and radius of localization increase which is probably triggered by Fe. However above 0.01, the rare earth element of

Eu became dominant. The 4f electrons from rare earth element will be bounded to the atomic core of samples, so that increasing the small amount of Fe did not affect to the *r* and μ in this system.

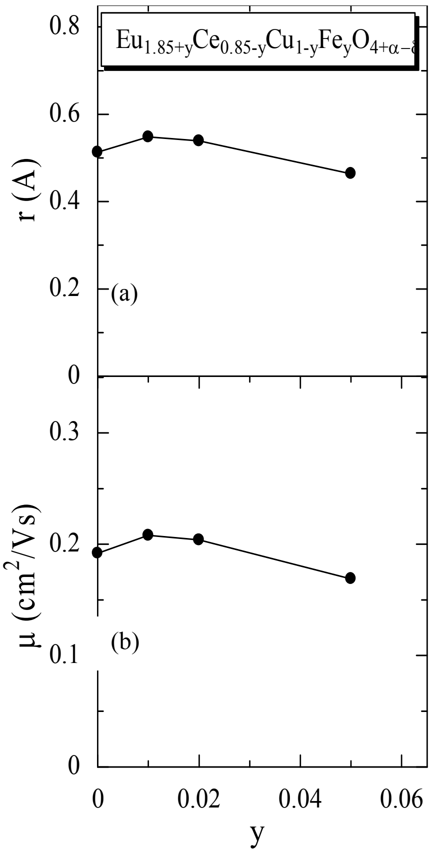

Fig. 5 The y value of concentration dependence of (a) radius of localization of electron (*r*), and (b) electron mobility (μ)

IV. Conclusion

Polycrystalline samples of $Eu_{1.85+y}Ce_{0.15-y}Cu_{1-y}Fe_yO_{4+\alpha-\delta}$ with y = 0, 0.010, 0.020, and 0.050 were prepared by ordinary solid-state reaction method in order to study the effect of partially substitution of magnetic impurity Fe for Cu to their magnetic and transport properties. The magnetic dc-susceptibility measurements show superconductivity with T_c onset about 15 K for y = 0 and completely disappear for y larger than 0.02 and 0.05. It is indicating that impurity Fe successfully disturbing spin-spin correlation in Cu layer. There is no significant changing of the value of Curie Constant and magnetic moment per atom per unit volume for y = 0.01. However, the value of C and $n \cdot m^2$ increase one order magnitude when y = 0.02 and 0.05 are applied that is probably due to the contribution of orbital moment of high spin of Fe to all magnetic moments in the samples.

Radius of localization of electron and electron mobility slightly increase for *y* = 0.01 which is probably triggered by Fe. However above 0.01, the rare earth element of Eu became dominant, so that increasing the small amount of Fe did not affect to the radius of localization and mobility of electron.

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