Magnetotransport properties and pseudogap phase diagram of superconducting EuBa$_2$Cu$_3$O$_y$ thin films: the influence of Eu substitution

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Abstract
This research explores and debates magnetotransport properties and the pseudogap phase diagram of high-temperature superconducting EuBa$_2$Cu$_3$O$_y$ (EBCO) films through \(\rho_{xx}(T)\) and \(\rho_{xy}(T)\) measurements. The mixed state transport properties of samples with higher doped levels show a power-law field-dependence of activation energy \(U, U \propto H^{-\alpha}\) with \(\alpha = 0.52 \pm 0.05\), while the slightly doped samples reveal a logarithmic field-dependence of \(U\). In addition, for the first time, all samples in the underdoped region display an unusual power-law behavior of \(U \propto p^\gamma\) with \(\gamma = -5.90 \pm 0.28\), which is qualitatively evaluated. A sign reversal of transverse resistivity in the mixed state is also present in the nearly optimum-doped samples, which have strong pinning energies. In the normal state, the cotangent of the Hall angle shows a wide range quadratic temperature dependence for all the samples, which can be described by Anderson’s theory. The values of spin excitation bandwidth \(W\) are coincident between two results obtained from the hidden Fermi-liquid theory and from the Hall angle for the slightly overdoped sample. Moreover, the phase diagram of EBCO, containing the superconducting transition temperatures, the \(T^*\) and \(T^{**}\) temperatures in the pseudogap (PG) phase, and the antiferromagnetic transition temperatures \(\theta_{\text{fl}}\) in the whole underdoped region has been completed and compared with those for YBa$_2$Cu$_3$O$_y$ (YBCO). The findings demonstrate that EBCO has lower \(T^*\) and \(T^{**}\) values, versus those of YBCO in the same doping region. The observed smaller magnitude of PG for EBCO is explained by considering the oxygen vibrational amplitudes in Cu$-$O$-$Cu bonds, which agrees with the inference based on the fluctuating bond model. The influence of Eu substitution is examined and discussed.

1. Introduction
The puzzling properties of magnetotransport have been at the center of debate in high-temperature superconducting (HTS) cuprates. Moreover, the microscopic origin of anomalous non-Fermi-liquid \(T\)-linear resistivity, the \(T\)-dependent Hall coefficient \(R_{\text{HH}}\), and the sign change of \(R_{\text{HH}}\) observed both in the normal state and the mixed state remain subjects of much discussion. Electrical resistivity measurements also reveal a remarkably \(T\)-linear behavior for samples near optimum doping, while a \(T^2\)-dependent feature can be observed in the higher-doping region [1, 2]. Thus, this study looks into the phase diagram of HTS cuprates, which is one of the most popular topics in condensed matter physics.

In the phase diagram, with an increase in the doping level, the highest critical temperature \(T_c\) of superconductors can be obtained at the optimally doped level. The superconducting phase boundary is generally considered as a parabolic-like curve empirically. In the higher-doping region, a line is usually drawn up and to the right from the edge of the superconducting dome in order to separate the non-Fermi-liquid (NFL) ‘strange metal’ from a conventional Fermi-liquid (FL) normal metal at high doping [2]. Recent studies on static magnetic susceptibility [3], the Nernst effect [4], neutron scattering [5], and transport coefficients [6] on underdoped HTS cuprates have revealed a possible normal state pseudogap (PG) in the phase diagram, which has been regarded as
the key to understanding superconductivity in cuprates [7]. The pseudogap phase occurs when the temperature decreases below a characteristic temperature $T^*$, called the pseudogap temperature. The opening of PG can be detected by a deviation in both the in-plane and c-axis resistivities from their linear-$T$ behavior at temperatures above the PG temperature $T^*$ [8]. Recent studies have argued that $T^*$ marks a phase transition into a state with broken time-reversal symmetry [9]. Some have also pointed out that the transport behavior in the PG state ($T_c < T < T^*$) is furthermore complicated due to possible superconducting, antiferromagnetic, and charge-spin stripe fluctuations [10, 11]. More recently, a simple $T^2$ dependence of resistivity was reported for underdoped HgBa$_2$CuO$_4$+$\delta$ and YBa$_2$Cu$_3$O$_{6.5}$ in the PG state, in which the in-plane magnetoresistance particularly obeys Kohler’s rule [12], thus inspiring scientists to conduct further studies on the phase diagram of high-$T_c$ superconductors.

EuBa$_2$Cu$_3$O$_7$ (EBCO) is a hole-doped cuprate superconductor with physical properties that are similar to the famous compound YBa$_2$Cu$_3$O$_7$ (YBCO). The lattice constants of the optimally doped EBCO are $a \approx 3.84$ Å, $b \approx 3.90$ Å, and $c \approx 11.70$ Å. As earlier reported on HTS REBa$_2$Cu$_3$O$_y$ (RE = rare-earth, except Ce, Pm, Tb, and Pr) cuprates, it has been shown that the crystal structure exhibits a tetragonal structure in the undoped region ($y < 6.4$), which becomes orthorhombic in the underdoped region [13, 14]. With a decrease of the oxygen content, the oxygen atom will be removed from the CuO chains, leading to strong doping dependences on many physical quantities. Therefore, EBCO also shows complex transport properties as observed on YBCO. Many studies regarding Y-substitution by trivalent RE ions have been conducted [15–17], and have shown that RE123 exhibits a systemic change of $T_c$ upon rare-earth substitution. However, the influence of the RE ionic radius on the crystal structure, the electron structure, and transport properties is not yet clear. Williams and Tallon [15] showed that, within the ionic bond-valence-sum model, the increased $T_c$ can be attributed to the increased hole distribution on the in-plane oxygen sites relative to the in-plane Cu sites, which accompany an increase in the c-axis coherence length and inter-plane coupling. Lütgemeier et al. [16] pointed out that the change of $T_c$ at increasing the ionic radii is also connected with differences in the order of oxygen. Recently Chen et al. [17] proved that the changes of the local electron density, the degree of orthorhombic distortion, and the coupling between the Cu–O chains and the CuO$_2$ planes, all have an effect on the superconductivity of RE123 cuprates. Thus, the nature of $T_c$ enhancement, as well as its further relation to the pseudogap and superconductivity, are now key issues for understanding RE123 cuprates. A great deal of effort has been made on the phase diagram of YBCO via resistivity, susceptibility, or high-energy x-ray diffraction studies [3–8, 18]. What seems to be lacking, however, is an investigation of the EBCO phase diagram.

This research prepared a series of EBCO samples with different hole doping levels so as to measure the magnetotransport properties. By changing the oxygen content in the EBCO thin films, the hole doping level varied from the optimum-doped region down to the nearly-insulated underdoped region. The in-plane longitudinal and transverse resistivities, $\rho_{xx}$ and $\rho_{xy}$, of the patterned EBCO thin films were measured from room temperature down to temperatures below $T_c$ in several fixed applied magnetic fields. Thus, this research presents the special temperature dependence of the resistivity of the cuprate superconductor and discusses an analysis of the results within the framework of studies on the phase diagram of HTS cuprates.

2. Experiments

Epitaxial c-axis-oriented EBCO thin films were deposited on (001) SrTiO$_3$ single crystal substrates by using the radio frequency (RF) magnetron sputtering method as used for YBCO thin films. Another study in the literature has offered a detailed description of the optimum sputtering conditions for the deposition of YBCO films [19]. Figure 1(a) shows a typical x-ray (Cu $K_\alpha$ radiation) diffraction pattern for an EBCO film with $T_c = 90$ K. As seen, only (00L) (L = 1–7) peaks are observed, indicating a strong c-axis orientation in the EBCO film. The as-deposited EBCO thin films in optimum sputtering conditions have $T_c$ values of around 91 K, with the superconducting critical temperature $T_c$ determined at zero resistance (within the noise level of $\pm 10^{-4}$ $\Omega$) as described elsewhere [20]. In order to achieve samples with different oxygen deficiencies, we treated the as-sputtered EBCO thin films with a re-annealing process. A high-temperature furnace with a quartz tube was used to anneal these deposited films for the purpose of removing oxygen atoms. The oxygen content of the EBCO thin films could be changed by thermal annealing below 500 °C without changing the stoichiometry of other elements [21]. Thus, the films were heated to different annealing temperatures of 150 °C–300 °C in a 1-atm argon atmosphere for 30 min to vary the oxygen contents in the films.

For the measurements of the transport properties, the EBCO films were patterned on Hall measurement geometry, which could be constructed to allow simultaneous measurements of both the longitudinal ($\rho_{xx}$) and transverse (Hall) resistivities ($\rho_{xy}$) using standard dc techniques. Hall voltages were taken in opposing fields parallel to the c-axis up to 6 T and at a dc current density of $\sim 1 \times 10^4$ A cm$^{-2}$. The magnetization was measured with a superconducting quantum interference device (SQUID) system (MPMS from Quantum Design).
3. Results and discussion

3.1. Longitudinal resistivity, Hall coefficient, and hole number

Figure 1 shows the temperature dependence of the zero-field longitudinal resistivity $\rho_{xx}$ for the samples studied. With the variation of the oxygen concentrations, the study obtained samples exhibiting different $T_c$ values of 4.5, 19.0, 53.8, 75.5, 65.8, 75.5, 84.8, 90.0, and 90.8 K, which can be denoted as S5, S19, S54, S66, S76, S85, S90, and S91, respectively. The inset of figure 1 shows the data for the two samples with lower oxygen contents, S5 and S19, which have lower $T_c$ values. The observed $T_c$ decreasing with a decrease in the oxygen content is consistent with that reported for YBCO [22].

Figure 2 shows the Hall coefficient $R_H$ versus temperature $T$ for all the samples with an applied magnetic field of 6 T along the crystalline $c$-axis. The positive sign of $R_H$ indicates a hole-dominating transport in EBCO. The Hall coefficient $R_H$ is directly related to the Hall carrier concentration ($n_H$) by the relation $R_H = \frac{1}{n_H e}$, and it can be found that the normal state $R_H$ increases for films with lower $T_c$, indicating that the carrier concentration fell with a decrease in $T_c$ (corresponding to a lower oxygen content in the sample). Furthermore, an obvious temperature dependence of $R_H$ can be seen in the samples with higher oxygen content, indicating that the Hall number $n_H$ is also strongly dependent on temperature.

It can be inferred that the Hall number increases with a rising temperature for the S54, S66, S76, S85, S90, and S91 samples, but the temperature dependence turns weaker for the S5 and S19 samples with lower oxygen content. These phenomena were also similar to those observed on YBCO as discussed in [23]. It can also be known that the primary effect of removing oxygen in HTS cuprates is to reduce the hole concentration $p$ on the CuO$_2$ planes. It has been universally agreed that there are many physical quantities of HTS cuprates which are highly dependent on the hole concentration $p$, which is defined as the number of holes per copper atom in the CuO$_2$ planes. Therefore, the determination of the $p$ value is a fundamental work for researching cuprate superconductors.

For the La$_{2-x}$Sr$_x$CuO$_4$ system, there exists a simple relation that $p$ is equal to the substitution concentration $x$. Due to this relation, the dependence between $T_c$ and $p$ can be approximately decided by the empirical parabola expression:

$$\frac{T_c}{T_{c,\text{max}}} = 1 - 82.6(p - 0.16)^2,$$

where $T_{c,\text{max}}$ is the $T_c$ of the optimally doped compound [24]. Equation (1) has been widely adopted to estimate $p$ for other HTS cuprates [25] when only considering the $T_c$ values. For a comparison between the measured Hall
number and the estimate $p$ according to equation (1), we redefine the $n_H$ value as the carrier number per copper atom, which can be calculated by:

$$n_H = \frac{V_{\text{cell}}}{N e R_H},$$

where $V_{\text{cell}}$ is the volume of a unit cell, and $N$ is the number of Cu atoms per unit cell. For EBCO, $N$ is equal to 2, and $V_{\text{cell}}$ is given by Tarascon et al\textsuperscript{[13]}.

Figure 3 shows the relationship between $p$ and $n_H$ taken at 290 K. Figure 3 also presents the reproduced $n_{H}(290 \text{ K})$ values of YBCO with different $p$ values reported by Cooper and Loram\textsuperscript{[26]} for comparison. One can see that the measured $n_H$ of EBCO is in agreement with that of YBCO, and the Hall numbers of both EBCO and YBCO with higher $p$ levels (i.e. higher $T_c$ values) deviate from the $n_H = p$ curve, which is derived from the $T_c$ values using equation (1). The inset of figure 3 demonstrates that the deviation between $n_H$ and $p$ indeed becomes larger for EBCO with higher $T_c$ values (i.e. a higher hole number in the CuO$_2$ planes). Particularly, the obtained $n_{H}(290 \text{ K})$ values of the EBCO films are nearly in coincidence with those for YBCO\textsuperscript{[26]}. As previously mentioned, equation (1) has been widely adopted to estimate $p$ for HTS cuprates, hence we chose the hole concentration $p$ instead of the Hall number $n_H$ at 290 K to describe the hole dopant in the samples and made the presented hole concentrations of EBCO films comparable to those previously reported.
3.2. Activation energy and mixed state Hall resistivity

A typical characteristic of type-II superconductors is that the samples can allow partial magnetic flux penetration in a mixed state. Transport properties in a mixed state have attracted significant attention since type-II cuprate superconductors were discovered. Figure 4 shows a typical temperature dependence of longitudinal resistivity $\rho_{xx}$ in a mixed state. Thermally activated flux flow behavior was first proposed by Anderson [30] and Kim et al [31] in early the 1960s and can be described by:

$$\rho_{xx}(T, H) = \rho_0 \exp (-U/k_B T),$$

where $U$ is the activation energy, and the prefactor $\rho_0$ is a resistivity regarded as not only temperature independent, but also field independent. We select the longitudinal resistivity at 100 K as the value of $\rho_0$ here.

The Arrhenius equation can be written equivalently as:

$$\ln \rho(T, H) - \ln \rho(H) = \frac{U}{k_B} \left( \frac{1}{T} \right).$$

The inset of figure 4 shows the Arrhenius plots of $\rho_{xx}(T)$ for the S91 sample in different fields. Thus, the activation energy $U$ can be obtained by the slope of a linear fitting line. Moreover, prominent magnetic field dependences of the activation energy $U$ appear in the experimental results. It can be observed that for most samples, $U$ can be fitted with a power-law dependence $U \propto H^{-\alpha}$, which has been widely followed by other type-II superconductors [32–34].

The log–log plots of the $U(H)$ curves seen in figure 5 demonstrate the power-law dependence for most EBCO thin films, and the values of $\alpha$ can be obtained by fitting. In figure 5 the dashed lines indicate the fitting of $U \propto H^{\alpha}$ with $\alpha = 0.52 \pm 0.05$ for the S91, S90, S85, S76, and S54 samples. Here, the presented $\alpha$ value is obtained by counting the average values of $\alpha$ for these five samples, which have close values of $\alpha = 0.48 \pm 0.03,$ $0.49 \pm 0.02,$ $0.54 \pm 0.04,$ $0.57 \pm 0.04$ and $0.53 \pm 0.05$, respectively. However, it is found that the $U(H)$ curves do not follow the power-law behavior for the slightly doped S19 and S5 samples. They exhibit logarithmic field-dependence, as shown in the inset of figure 5. This result is likely to be the same as that observed on YBa$_2$Cu$_3$O$_y$/PrBa$_2$Cu$_3$O$_y$ superlattices proposed by Brunner et al [35]. In addition, one can see that the samples with a near optimum doping level exhibit strong vortex pinning and have the same order of magnitude of $U$ at about $10^4$ K as that of YBCO [33]. In addition to the different $U(H)$ behaviors, figure 5 also shows an extremely wide distribution of $U$ values for the samples, ranging from 39 000 to 19 K respectively for S91 and S5 at 0.25 T, for example. This implies that the activation energy is strongly dependent on hole concentration $p$ for the cuprate superconductors.

Furthermore, since the activation energy is determined at different reduced temperatures, it is necessary to take account of the temperature and field dependences. A modified method to calculate the activation energy from the resistance transition has been proposed [36, 37]. Following the procedures given in [36, 37], we extract the temperature dependence of $U$ with a choice of $U(t) = U(H)(1 - t)^m$, where the reduced temperature $t = T/T_c(H)$, $U^*(H)$ corresponds to the activation energy at 0 K, and $m = 2$ as given in this modified method. Here $T_c(H)$ is the mean-field transition temperature which can be estimated from the resistive transition curves.
in fields. In addition, we retained the unknown field-dependence of $U^*(H)$ to simplify the fitting procedures. We replotted the logarithmic data in figure 4 as a function of $(1 - \frac{t}{T})^2$ and obtained a straight line behavior to determine the $U^*$ value. The values of exacted activation energy $U^*$ for S91 in different fields are also shown in figure 5 for comparison. As seen, the zero-temperature $U^*$ is reasonably much larger than that determined from the simple Arrhenius plot, $U$, and also shows a power-law dependence $U^* \propto H^{-\alpha}$ with $\alpha = 0.93 \pm 0.02$, which is near unity as previously reported [37]. The different $\alpha$ values in the power-law dependences of $U$ and $U^*$ are possibly due to the different temperature regions extracted from the $\rho(T)$ curve. Since similar field-dependent behaviors of $U$ and $U^*$ were observed, we chose the values of $U^*$ for the following discussions.

Figure 6 shows the hole concentration $p$ dependence of $U$ for the samples under applied fields of 0.5–6 T. Note that the vertical axis is a logarithmic scale ranging in the order of a magnitude of five, indicating a drastic variation of $U$ for samples with different hole doping levels. In other words, the activation energy $U$ decreases rapidly with a reduction of the oxygen concentration in the EBCO films. It is found that the relationship between $U$ and $p$ can be described by a power-law fitting of $U \propto p^\gamma$ with $\gamma = -5.90 \pm 0.28$ in the EBCO films, where the negative value of $\gamma$ indicates the increase in $U$ with an increase of $p$, as the $p$ value is smaller than one. Surprisingly, little attention has been given to this interesting observation. Recently, Naqib and Islam [37] showed a greatly significant effect of doped hole number $p$ on the activation energy $U$ in YBCO films via changing the oxygen deficiency in the Cu–O chains, as conducted by this study. They demonstrated that the dimensionless activation energy can be expressed as $U(T, H) = (1 - \frac{t}{T})\ln(H_0/H)^\beta$, where $t$ is the reduced temperature and $H_0$ is a characteristic field scale. As shown in their study, the value of temperature exponent $m$ depends on the hole content with an approximate dependence of $m \propto p$, whereas $\beta$ is insensitive to the $p$ values and is close to unity. In addition, $H_0$ decreases rapidly as $p$ is reduced, i.e., $H_0$ is also approximately proportional to $p$. Considering both $t$ and $H_0$ dependences, a strong $p$ dependence of $U$, $U \propto p^\gamma$ with $|\gamma| \gg \beta$, can be
predicted. For the first time, the strong $p$ power-law dependence of $U$ is presented for HTS cuprates, which can be qualitatively explained.

On the other hand, the Hall effect in the mixed state of type-II superconductors has been one of the most interesting subjects of the past two decades and remains a puzzle to researchers. The anomalously temperature-dependent behavior of the mixed state Hall resistivity $\rho_{xy}$ reveals a remarkable sign reversal of $\rho_{xy}$ near superconducting transition, which is not only a phenomenon near $T_c$, but a characteristic of the mixed state. Many researchers have argued that this effect should be viewed as a general consequence of vortex dynamics and is probably caused by flux pinning \[38–40\]. In the mixed state Hall measurement on the EBCO thin films, the sign reversal of $\rho_{xy}$ can be observed in the two highest $T_c$ samples.

Figures 7(a) and (b) show two typical temperature dependences of Hall resistivity $\rho_{xy}$ in fields of 1, 3, 5, and 6 T for the S90 and S85 samples, respectively. It can be clearly observed that sample S90 exhibits the sign reversal of $\rho_{xy}$, while sample S85 does not go into a sign reversal in the mixed state. The other lower-doped samples also do not exhibit any sign reversal of $\rho_{xy}$ either. This result is similar to that for YBCO single crystals proposed by Jin and Ott \[41\]. A general explanation is that a strong vortex pinning can bring about the sign reversal of $\rho_{xy}$ in the mixed state \[38\]. Inspecting the fact that the activation energy of S90 is larger than that of S85, we can recognize that the samples with stronger vortex pinning indeed exhibit this anomalous phenomenon.

### 3.3. Normal state Hall angle and spin exchange energy

The normal state transport properties of HTS cuprates have revealed many noticeable characteristics, in which the anomalous temperature dependences of the longitudinal and transverse (Hall) resistivities, $\rho_{xx}$ and $\rho_{xy}$, in the $ab$ plane still present some puzzles up to now. In the normal state, one of the most remarkable properties of many HTS cuprates is that the cotangent of the Hall angle $\cot \theta_H = \rho_{xx} / \rho_{xy}$ shows a wide range quadratic temperature dependence. Anderson’s theory explained this phenomenon and indicated that the transport relaxation rate $1/\tau_1$ for resistivity is proportional to $T$, and that the transverse (Hall) relaxation rate $1/\tau_H$ is proportional to $T^2$ caused by scattering between spin excitations alone \[42, 43\]. By considering the scattering rate of $1/\tau_M$ due to magnetically active impurities, the transverse (Hall) scattering rate can be written as:

$$
\frac{1}{\tau_H} = \frac{T^2}{W} + \frac{1}{\tau_M},
$$

where $W$ is the bandwidth of the spin excitation and corresponds to spin exchange energy $J$ in Anderson’s theory. By the definition of the Hall angle, the cotangent of the Hall angle $\cot \theta_H$ involves $1/\tau_H$ only. Equation (5) implies that:

$$
\cot \theta_H = \Lambda T^2 + C^*,
$$

where $C^*$ is the magnetic impurity contribution. This equation shows that the cotangent of the Hall angle $\cot \theta_H$ exhibits quadratic temperature dependence, which has been observed in many cuprate superconductors \[43–45\].
To newly describe the temperature dependence of longitudinal resistivity, some theoretical works consider that there are two regions; one is the linear-$T\rho_{xx}$ in the strange metal region, and the other is $T^2$-dependent $\rho_{xx}$ for lower temperature and higher doped cuprates [46, 47]. For quantitative analyses, Anderson and Casey proposed the hidden Fermi-liquid theory and pointed out that there is no clear boundary between the two regions [46]. The anomalous behavior of the temperature dependence of the longitudinal resistivity in the normal state is described by the hidden Fermi-liquid theory [48] and can be denoted as:

$$\rho(T) = \frac{\hbar}{e^2E_F} \frac{T^2}{T + W} + \rho_{\text{res}},$$

(7)

where $E_F$ is the Fermi energy, $W$ is the bandwidth, and $\rho_{\text{res}}$ is the residual resistivity. The parameter $W$ plays a crucial role in this equation, because it determines the temperature-dependent behavior of longitudinal resistivity $\rho_{xx}$. It can be found that if the $W$ is small, the resistivity will show linear temperature dependence. If $W$ is about several hundred degrees Kelvin, then the resistivity at low temperatures will obey the $T^2$ law, but it will show approximately linear temperature dependence at the higher temperature region.

Equation (7) can be reduced to the following form [48]:

$$\frac{d[\ln(\rho - \rho_{\text{res}})]}{d[\ln T]} = 1 + \frac{W}{T + W}.$$  

(8)

Thus, the prefactor in equation (7) can be canceled, and it becomes easy to fit the resistivity as a function of temperature due to eliminating a parameter. The results of fitting by the hidden Fermi-liquid method have been confirmed in the data of both polycrystalline and single crystal La$_{2-x}$Sr$_x$CuO$_4$ and show that $W$ increases with a rising doping level in the overdoped region [48–50]. Another work proposed by Anderson and Casey provides a relationship for $W$ and $\lambda$ to prove the theory is self-consistent [51], in which $W$ can be deduced from the Hall angle by:

$$W = \sqrt{\frac{n_2\hbar}{\Lambda eB}}.$$  

(9)

Here $n$ is the two-dimensional carrier density, which is counted by $n_{2D} = n_{3D} \times 11.71 \text{ Å}/2$, where 11.71 Å is the $c$-axis lattice constant. The factor of 2 means that there are two CuO$_2$ layers in an EBCO unit cell, while $\lambda$ is the slope of $\cot \theta_H$ versus $T^2$ in equation (6), and $B$ is the applied magnetic field.

According to equation (9), $W$ for Tl$_2$Ba$_2$CuO$_{6+x}$ was calculated using the $\cot \theta_H$ data and yielded $W = 800$ K, which was consistent with the fitting result from the resistivity using equation (8) [50–52].

Figure 8(a) shows the results of the fit for the four highest $T_c$ EBCO samples, S91, S90, S85, and S76, in the normal state using equation (8). Figure 8(b) presents the results of $\cot \theta_H$ versus $T^2$ for the corresponding samples under the applied magnetic field of 6 T. As seen, the data of samples with different doping levels almost fall on a straight dash line and can be fitted to equation (6). It is noteworthy that, an early study [13] reported a much larger normal state magnetic susceptibility of EBCO than those for other RE123 compounds. It has been
pointed out that Europium is one of the few rare earths that can exist under two oxidation states (2+ and 3+), and that Eu can act as a magnetic impurity on Ba sites, because Eu2+ is magnetic. Hence, EBCO is a good candidate to clarify Anderson’s theory. Indeed, the obtained average value of $C^*$ is around 46.4 for the EBCO films, which is much larger than those of 13 and 5 reported for YBCO films and single crystals, respectively [43, 53]. This result confirms Anderson’s theory in explaining the normal state Hall behavior, with the conjecture that there exist two transport relaxation times that independently influence the Hall effect and resistivity in the cuprates. However, it should be noted that the presence of mixed Eu valence seems to be a problem since the Eu2+ may increase the hole content in comparison with YBCO. To debate this problem, we can recall the $n_{ff}$ data shown in figure 3, in which it can be seen that the measured $n_{ff}$ of EBCO is in agreement with that of YBCO. This result indicates that even though the mixed valence of Eu exists, the Eu2+ ions do not exhibit a significant contribution to the hole content in the CuO$_2$ plane. Thus, it can be inferred that the extra hole numbers due to the presence of Eu2+ ions are localized and the conductive in-plane hole carriers are dominated by the oxygen content in the EBCO films. The inset of figure 8(b) shows the analytical results of W derived from equations (8) and (9) versus the hole concentration $p$. By using equation (8), it can be seen that the derived W values of the three lower doping samples, S90, S85, and S76, are approximately 10 K and are one order of magnitude smaller than the values (∼200 K) calculated by equation (9). The data of W are clearly inconsistent between two results respectively obtained from the hidden Fermi-liquid theory and Hall angle for samples in the underdoped region.

Turning now to the $W$ data of the S91 sample, for the sample with highest $T_c$ value, however, it can be seen that the resistivity behavior shows a slightly upward curve as shown in figure 8(a), implying that the film is possibly a slightly overdoped sample. The value of fitting parameter $W$ from equation (8) is 262 K, which is close to the $W$ value of 315 K obtained from equation (9). This result for the S91 sample is in accordance with that for overdoped Tl$_2$Ba$_2$CuO$_6$+$\delta$ [51]. The inference of the slightly overdoped S91 can also be confirmed by the $n_{ff}$ result, which shows a larger $n_{ff}$ value for S91 than those of the other samples, as seen in the inset of figure 3. Moreover, the inconsistency between the two results of $W$ respectively obtained from equations (8) and (9) for the underdoped samples as well as the discontinuous transition of $W$ to 262 K of S91, as shown in the inset of figure 8(b), can be understood according to the reported fact that the values of W obtained from equations (8) and (9) are consistent only on the overdoped samples [48, 51]. The normal state resistivity behavior in the strange metal region remains a matter for debate; nevertheless, our analysis can provide a reference for this new theory in an attempt to understand the enigma of HTS cuprates.

### 3.4. Pseudogap phase diagram

One of the greatest puzzles of high-$T_c$ superconductors is the PG region in the cuprate phase diagram, as mentioned previously. We attempted to define the PG temperature $T^*$ from the electrical resistivity measurement, which has been considered one of the most useful methods. Figure 9(a) shows the $\rho_{xx}(T)$ curves for three typical underdoped samples, S54, S19, and S5. As seen, the $\rho_{xx}(T)$ curves exhibit a downward deviation from the linear behavior, and $T^*$ is defined as the temperature where resistivity starts to decrease below such linearity. Here, $T^*$ is determined by the $\frac{1}{4}$% deviation of the $\rho_{xx}(T)$ curve from the linear fitting in the high-temperature regime. However, it is difficult to determine the PG temperature $T^*$ directly from the $\rho_{xx}(T)$ curves for these samples in approximately the optimum-doped region, in which a wide range of $T$-linear behavior can be observed as previously seen in figure 1. This problem can be resolved by plotting the data as $[\rho_{xx}(T) - \rho_{xx}(0)]/\beta T$ versus $T$, as shown in the inset of figure 9(a), where $\beta$ is the slope of the $T$-linear region of the $\rho_{xx}(T)$ curve, and $\rho_{xx}(0)$ is the residual resistivity defined as the intercept of the line extrapolated from the $T$-linear region at $T = 0$ K.

In the PG regime a quadratic temperature dependence was first reported for overdoped HgBa$_2$CuO$_{4+x}$ [54], in which the resistivity shows a Fermi-liquid-like behavior below a characteristic temperature $T^\ast\ast$ in the normal state as mentioned previously. This phenomenon also appears in some slightly doped cuprates, such as YBa$_2$Cu$_3$O$_{6+y}$ [55] and La$_2$-Sr$_x$CuO$_4$ [56]. In this study, quadratic resistive behavior in EBCO was observed for only the three lowest $T_c$ samples, S54, S19, and S5. Figure 9(b) shows that the resistivity exhibits a quadratic temperature dependence below $T^\ast\ast$ for the S54, S19, and S5 samples. The dashed lines fit the $T^2$ dependence range, and $T^\ast\ast$ is determined also by the $\frac{1}{4}$% deviation of the $\rho_{xx}(T)$ curve from the $T^2$ dependence as marked by an arrow. The determination methods of $T^*$ and $T^\ast\ast$ are similar to those reported in articles [6, 12, 54].

We attempt to complete the phase diagram for EBCO in the extremely low-$p$ region, in which an antiferromagnetic (AF) transition has been predicted as seen for YBCO [18]. Three EBCO polycrystalline samples, denoted by S003, S004, and S005, have been prepared by the solid-state reaction method and annealed at 460 °C–480 °C in an argon atmosphere for 5–15 h to vary the oxygen contents (as seen in table 1). Temperature dependences of the magnetization for bulk samples were measured, and then the samples were grounded for powder x-ray diffraction measurement. The AF transition temperature (related to the Néel
temperature) \( \theta_N \) can be determined by the magnetization measurement, following the temperature dependence of \( M = C/(T + \theta_N) \), where \( C \) is the Curie constant.

Figures 10(a)–(c) show \( 1/M \) as a function of temperature for samples S003, S004, and S005, respectively. The values of Néel temperature \( \theta_N \), extrapolated from the linear temperature dependence of \( 1/M \) in the paramagnetic state, are 312 \( \pm \) 1, 288 \( \pm \) 3, and 272 \( \pm \) 2 K for samples S003, S004, and S005, respectively. Having observed the AF transition, we then considered the determination of hole concentration \( p \) in these polycrystalline samples. Unfortunately, the S003, S004, and S005 samples exhibit an insulator characteristic where no superconducting transition is observed, leading to the uselessness of equation (1). Thus, another way to determine \( p \) for such insulating cuprates is invoked.

Liang et al proposed a way to determine \( p \) using the c-axis lattice constant for YBCO [57]. The inset of figure 10(a) shows the x-ray diffraction (007) peaks for the S003, S004, and S005 samples, which correspond to the c-axis lattice constants of 1.1784, 1.1776, and 1.1768 nm, respectively. Following the work of Liang et al, we can determine the \( p \) values based on the presented linear relationship between \( p \) and \( c \) in the low-doping region. The two reference points for the linear fitting line were chosen at \( p = 0, c_0 = 1.183 \) nm and \( p = 0.05, c = 1.1764 \) nm. The \( c_0 \) was obtained by using the two EBCO c-axis lattice constants reported in [58] and extrapolating to that of EBCO with \( y = 6 \) [39]. The second reference point came from another powder EBCO sample with \( T_c \leq 2 \) K (the lowest temperature in the SQUID system). Thus, the obtained \( p \) values are 0.033, 0.040, and 0.047 holes/Cu for the S003, S004, and S005 samples, respectively. It is worth mentioning that we did

| Samples | \( T_a \) (°C) | \( T_c \) (K) | \( \theta_N \) (K) | \( T^* \) (K) | \( T^{**} \) (K) | \( p \) (holes/Cu) | \( U(3 \text{ T}) \) (K) | \( A \) (K^{-1}) |
|---------|---------------|--------------|----------------|-------------|----------------|-----------------|----------------|------------|
| S91     | —             | 90.8         | —              | —           | —              | 0.165           | 11 908         | 0.0080     |
| S90     | 190           | 90.0         | 146 ± 14       | —           | —              | 0.148           | 7486           | 0.0125     |
| S85     | 200           | 84.8         | 156 ± 10       | —           | —              | 0.131           | 3306           | 0.0150     |
| S76     | 230           | 75.5         | 156 ± 6        | —           | —              | 0.115           | 1489           | 0.0121     |
| S66     | 235           | 65.8         | 163 ± 12       | —           | —              | 0.102           | 1135           | 0.0120     |
| S54     | 235           | 53.8         | 208 ± 6        | 125 ± 4     | 0.090          | 538             | 0.0108        |
| S19     | 250           | 19.0         | 252 ± 2        | 145 ± 4     | 0.062          | 68              | 0.0075        |
| S5      | 260           | 4.5          | 286 ± 2        | 170 ± 4     | 0.053          | 10              | 0.0139        |
| S005    | 460           | —            | 272 ± 2        | —           | —              | 0.047           | —              | —          |
| S004    | 470           | —            | 288 ± 3        | —           | —              | 0.040           | —              | —          |
| S003    | 480           | —            | 312 ± 1        | —           | —              | 0.033           | —              | —          |
not use the \( c \)-axis lattice constant to determine the \( p \) values for the thin-film samples (S5, S19, S54, etc) owing to the strain effect on the crystalline structure. For the moment, we are ready to extend these observations of \( T_c \), \( T^* \), \( T^{**} \), and \( \theta_N \) into the idea of a completed EBCO phase diagram.

Before addressing the inquiry of the lower \( T^* \), as well as the \( T^{**} \) values for EBCO films, one point of the twinned structure effect in films requires clarification. Several oxygen-deficient YBCO thin-film samples were fabricated by the same RF sputtering method, which were studied to compare with the results of the EBCO films and SC YBCO samples. Figure 11 shows a typical comparison of the resistive transition between a YBCO film and an EBCO film, with \( T_c = 69 \) and 66 K, respectively. It can be seen that the \( T^* \) value of the EBCO film is obviously lower than that of the YBCO film, even though both reveal a near superconducting transition temperature \( T_c \). The inset of figure 11 illustrates the \( T^* \) boundaries of the YBCO films and SC YBCO samples for comparison. It can be seen that our twinned YBCO films have nearly the same PG boundary as those of the SC
YBCO samples given by other groups, indicating that the PG boundary is not significantly affected by the twinned structure commonly seen in thin-film samples. It is worth mentioning that, the obtained $T^*$ of $154 \pm 7$ K for our optimally doped 90 K $T_c$ YBCO films is close to the reported 152 K for the 93 K $T_c$ YBCO film grown by laser molecular beam epitaxy deposition [60]. The EBCO phase diagram has been completed with the modified $p$ values, as shown in figure 12. Table 1 lists the parameters, including the annealing temperature $T_a$, $T_c$, $\theta_N$, $T^*$, $T^{**}$, $p$, $U$, and $A$, as obtained in this work, for all samples. As seen in figure 12, the temperature-doping phase diagram for EBCO shows that both $T^*$ and $T^{**}$ decrease with an increasing doping level $p$, and $\theta_N$ falls drastically to the lowest point at $p \approx 0.05$ holes/Cu. The trend shown in the phase diagram for EBCO is consistent with those for other cuprates [18, 54]. The values of $T_a$, $T_c$, and $T^*$ for the SC YBCO samples, as reproduced from other studies [4, 12, 54, 57] are also shown in figure 12 for comparison. It can be seen that both the underdoped EBCO and YBCO samples reveal the characteristic temperatures that exhibit $T^* > T^{**} > T_c$ in the phase diagram. Here, the $T_c$ of EBCO almost merges with that of YBCO presented in [57]. However, it is found that EBCO demonstrates lower values of $T^*$ compared to those of the SC YBCO samples in the same doping region, where the $T^*$ values of YBCO were obtained by the Nernst effect [4] and the deviation in the in-plane linear-T resistivities [54], respectively. Moreover, the $T^{**}$ of EBCO also shows lower values compared to those of YBCO [12, 54] in the same doping region.

Returning to the real subject of the lower PG temperatures observed on EBCO films, it is well known that the difference between EBCO and YBCO is that the position of Y is substituted by the larger Europium atom. This atom is at the center of one unit cell, and separates the two electrical-transport CuO$_2$ layers, leading to a change in superconductivity, as previously mentioned [15–17]. There are many theories that try to explain the mechanisms of the PG phase and high-temperature superconductivity; however, little attention has been given to this point regarding the RE ion substitution. It has been proposed that, upon reaching $T^*$, a gap opens in the hot quasiparticle spectrum, thus, establishing the PG, while superconductivity occurs when the cold quasiparticles become gapped [7] and are independent of the hot quasiparticles that are already gapped at $T_c$. Thus, it is argued that the magnitude of PG and $T^*$ are unrelated to $T_c$. Indeed, a recent study on Zn substituted La214 cuprates [61] has shown that Zn acts as an efficient suppressor of $T_c$, but has no effect on PG for a given hole content. It is noteworthy that both PG and the enhancement of magnetic susceptibility increasing in magnitude with increasing underdoping are presented in this study. The results indicate that the variations of $T^*$ as well as $T^{**}$ can be due to magnetic disorders induced in the in-gap states in the underdoped region. Recently, many articles have given a basic picture, namely, the two-dimensional Hubbard model capture, to describe the basic physics of superconductivity and the pseudogap, as seen in [62–64], as well as other references herein, for a comprehensive overview. On the other hand, a series of theoretical approaches based on anharmonic oxygen degrees of freedom, and termed as the fluctuating bond model (FBM), have also attempted to describe the phase diagram of HTS cuprates [65, 66]. This microscopic theory treats the vibrational degrees of freedom of CuO$_2$ planar oxygen transverse to the Cu–O–Cu bond, as well as their interaction with the electron system in an anharmonic model. The theoretical approach is based on experiments of low temperature scanning tunneling microscopy spectroscopy [67], which show the PG phase boundary at $T^{**}$ as a symmetry breaking from a C4 symmetry above $T^*$ to C2 below $T^*$. However, in view of these theoretical approaches, the lower $T^*$, as well as $T^{**}$ values for EBCO, seemingly remain a puzzle. Upon interpretation, it is worth noting that, in REBa$_2$Cu$_3$O$_{7-\delta}$ compounds with an increasing RE ion size, the difference between the in-plane $a$-axis and $b$-axis lattice constant, $|a-b|$, is decreased, while the buckling of the Cu–O planes is almost independent of the RE ion.

![Figure 12. Completed EBCO phase diagram with $p$ values involving the $T_c$, $T^*$, $T^{**}$, and $\theta_N$ parameters in the diagram. The values of $T_c$, $T^*$, and $T^{**}$ for YBCO reproduced from other studies are also shown for comparison. The dashed lines for the EBCO $T^*$ and $T^{**}$ data are for the purpose of guiding the eye.](image)
size [68, 69]. Within the framework of FBM [65], the magnitude of PG is calculated by considering the differences of the oxygen vibrational amplitudes or the buckling angles between the $a$-aligned and $b$-aligned Cu–O–Cu bonds, i.e., $|\langle u_a^2 \rangle - \langle u_b^2 \rangle|$, where $u_a$ and $u_b$ are the oxygen distance from the $a$-aligned and $b$-aligned Cu–O–Cu bond axes, respectively. The $u_a$ value, for example, can be estimated by $u_a = \frac{a}{2} \cot (\phi_a/2)$ with buckling angle $\phi_a$ in the $a$-aligned Cu–O–Cu bond [70]. Therefore, we can roughly calculate the values of $|\langle u_a^2 \rangle - \langle u_b^2 \rangle|$ for EBCO and YBCO according to the reported structure data that $a = 3.836(3.817) \, \text{Å}$, $b = 3.897(3.883) \, \text{Å}$, $\phi_a \approx 163.4^\circ (163.8^\circ)$, and $\phi_b \approx 164.4^\circ (164.9^\circ)$, for EBCO (YBCO) [69]. The obtained value of $|\langle u_a^2 \rangle - \langle u_b^2 \rangle|$ for EBCO is $\approx 7.049 \times 10^{-3} \, \text{Å}^2$, which is smaller than that of $7.560 \times 10^{-3} \, \text{Å}^2$ for YBCO. Thus, the observed lower $T^*$ values in the EBCO films are undoubtedly in accordance with the inference based on the FBM theory. The phase diagram of EBCO is presented and discussed in this theory, for the first time, in order to offer the key to understanding high-temperature superconductivity.

4. Conclusion

This study has explored magnetotransport properties and the PG phase diagram of HTS EBCO films grown by the RF sputtering technique. The values of $T_c$ and doping level $p$ for EBCO films can be controlled via the processes with different post-annealing conditions. Magnetotransport properties of the samples were debated using the $\rho_{\alpha\beta}(T)$ and $\rho_{xx}(T)$ measurements. It is observed that the measured Hall numbers for EBCO with higher $p$ levels (higher oxygen contents and higher $T_c$ values) deviate from the $p$ values derived using equation (1). Inspecting the mixed state transport properties, it is found that the relation between $U$ and $H$ can be well described by the power-law dependence of $U \propto H^{-\alpha}$ with $\alpha = 0.52 \pm 0.05$ in the samples with higher doped levels, while the slightly doped samples show logarithmic field-dependence. In addition, an unusual power-law behavior of $U \propto p^\gamma$ with $\gamma = -5.90 \pm 0.28$ is displayed and qualitatively explained, for the first time, for all samples in the underdoped region.

Sign reversal of transverse resistivity in the mixed state is also observed in the nearly optimum-doped samples, which have strong pinning energies. This observation is consistent with the prediction in theory. In the normal state, the cotangent of the Hall angle shows a wide range of quadratic temperature dependence for all the samples and has been explained by Anderson’s theory. However, the data of $W$ are coincident between the two results respectively obtained from the hidden Fermi-liquid theory and the Hall angle only for the slightly overdoped sample, S91. Moreover, the phase diagram of HTS EBCO, containing the superconducting transition temperatures, the $T^*$ and $T^{**}$ temperatures in the PG phase, and the AF transition temperatures $\theta_{\alpha\beta}$ in the whole underdoped region, has been completed and compared with those for YBCO. In particular, it is found that EBCO demonstrates lower values of $T^*$ and $T^{**}$ compared to those of YBCO in the same doping region. This puzzling result has been clarified within the framework of the FBM theory. Considering the reported structure data, the estimated magnitude of PG for EBCO is smaller than that of YBCO, which is in agreement with the inference based on the FBM theory. The phase diagram of EBCO is presented and discussed to further recognize high-temperature superconductivity.

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