Normal-state transport properties of $YBa_2Cu_3O_{7-\delta}/PrBa_2Cu_3O_{7-\delta}$ superlattices

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The experimental results of resistivity and thermoelectric power (TEP) of two $YBa_2Cu_3O_{7-\delta}/PrBa_2Cu_3O_{7-\delta}$ (YBCO/PrBCO) superlattice samples with different thickness of YBCO layers in the temperature range from 80 to 270 K are reported. The contribution of interface scattering to the resistivity of YBCO layers is estimated. Compared with slightly oxygen-deficient YBCO films, the TEP of superlattice samples has higher positive value and the slope of the linearly temperature-dependent part changes from negative to positive. A TEP peak just above T_c was also observed.

I. INTRODUCTION

Recently, the structure and properties of the $YBa_2Cu_3O_{7-\delta}/PrBa_2Cu_3O_{7-\delta}$ (YBCO/PrBCO) superlattice attracted much attention not only because it might shed new light on the studies of the layered materials but also because of the characteristics of the superlattice itself.¹⁻¹⁰ The transport properties in the normal state of the superlattice can provide us information about the sign and concentration of carriers, the electronic band structure, and the transport mechanism. However, most work on this aspect, both experimental¹⁻³ and theoretical, $^{8-10}$ has been focused on the properties of the transition from the superconducting state to the normal state. Only a few groups reported on the transport properties in the normal state with brief discussions. 6,7 In this paper, we present our experimental results of the resistivity and thermoelectric power (TEP) measurements on the YBCO/PrBCO superlattice from 80 to 270 K.

II. EXPERIMENTAL DETAILS AND RESULTS

The samples were grown by laser ablation onto singlecrystal $Zr(Y)O_2$ substrates. Layers of $PrBa_2Cu_3O_{7-\delta}$ and of $YBa_2Cu_3O_{7-\delta}$ were deposited alternatively with the total thickness of about 200 nm. The surface layer is YBCO. X-ray-diffraction measurement showed that the *c* axis is oriented perpendicular to the substrate surface and satellite peaks were clearly observed. Two superlattice samples have been studied, in which the individual PrBCO layer thickness was chosen to be 3.6 nm (three unit cells) and the individual YBCO layer thickness was 8.4 and 6.0 nm for samples *A* and *B*, respectively. For comparison, $YBa_2Cu_3O_{7-\delta}$, $PrBa_2Cu_3O_{7-\delta}$, and $Y_{0.8}Pr_{0.2}Ba_2CuO_{7-\delta}$ (YPrBCO) films with thickness of about 200 nm were also prepared.

The resistivity was measured by the standard fourprobe method. The TEP was measured using the differential method and corrected for the contributions from both the copper leads and the spurious voltage. Electrical contacts to the sample were made by coldpressed indium pads.

The resistivity $\rho(T)$ for the superlattice sample A and a

YBCO film are shown in Fig. 1. For the YBCO film, the result is typical, with linear temperature dependence above T_c and values ranging from 100-300 $\mu\Omega$ cm.¹¹ While the resistivity in the superlattice sample has different characters: (1) The values, being about 400-800 $\mu\Omega$ cm above T_c , are larger than that of the YBCO film. (2) Though it increases linearly with temperature above T_c , there is a deviation from the straight line in the $\rho(T)$ curve above 200 K. Similar results were obtained by Kerchner *et al.*⁶

The measured thermoelectric power S(T) is shown in Fig. 2. Slightly oxygen-deficient YBCO samples usually have positive TEP value and a negative slope dS(T)/dT,¹²⁻¹⁴ as shown in Fig. 2 for one of our YBCO films. The results of the YBCO/PrBCO superlattice samples are different from that of the YBCO film in the following aspects: (1) The value is larger, which is about $10-20 \mu V/K$ above T_c . (2) It varies linearly with temperature, but has a positive instead of a negative slope. In addition, there is a broad peak just above T_c in the S(T) curve of sample A. To the best of our knowledge, this is the first measurement of TEP on the



FIG. 1. Resistivity of YBCO/PrBCO superlattice and YBCO film. The dotted line is the fit to the data using Eq. (1).



FIG. 2. Thermoelectric power of YBCO/PrBCO superlattice, YBCO film, and YPrBCO film. The dotted line is the fit to the data using Eq. (2).

YBCO/PrBCO superlattice. The TEP of a YPrBCO film is also shown in Fig. 2.

The resistivity and TEP of PrBCO film are shown in Figs. 3(a) and 3(b). The resistivity of PrBCO is very large $(10^4-10^6 \ \mu\Omega \ cm)$ and decreases with increasing temperature, which shows semiconductorlike behavior. The TEP of PrBCO has large value (20-100 μ V/K) with positive temperature coefficient. All of these are consistent with reported results.^{15,16}

III. DISCUSSION

From results mentioned above, it can be seen that the TEP behavior of the YBCO/PrBCO superlattice is totally different from that of YPrBCO (Fig. 2). The difference is also shown in the resistivity and Hall-effect measurements.^{7,17} Thus the distinctive transport properties of the YBCO/PrBCO superlattice in the normal state do not result from the stoichiometric diffusion between the YBCO layers and PrBCO layers. Investigation of our samples and others by Z-contrast TEM (transmission electron microscopy) revealed that the interfaces are very clear and no evidence of the stoichiometric diffusion between the YBCO layers and PrBCO layers.⁵

The contacts made to the superlattice samples in our case may lead to a current component perpendicular to the layers and consequently an inhomogeneous current distribution inside the sample. Busch et al. have disnonuniform current distribution cussed the in $Bi_2Sr_2CaCu_2O_x$ single crystal within an anisotropy resistivity model.¹⁸ Extending their model to our case and considering that the voltage contacts, near the middle of the sample, are far away from the current contacts at the edge and, particularly, the ratio of the sample thickness to its length in our case is about 10^{-5} , three orders of magnitude smaller than that in Busch's work, we believe that the measured resistance in our work is a parallel resistance of the alternately arranged layers. Therefore,

the results are analyzed by a parallel channel model, which is the YBCO layers and PrBCO layers are electrically parallel connected in the superlattice sample. If the properties of YBCO and PrBCO layers in the superlattice have no difference with that of bulk samples, using the measured resistivity and TEP of the YBCO film, $\rho_{\rm Y}$, $S_{\rm Y}$, and that of PrBCO film $\rho_{\rm Pr}$, $S_{\rm Pr}$, the resistivity and the TEP of the superlattice samples, $\rho_{\rm SL}$, $S_{\rm SL}$ can be calculated as¹⁹

$$\frac{d}{\rho_{\rm SY}} = \frac{d_{\rm Y}}{\rho_{\rm Y}} + \frac{d_{\rm Pr}}{\rho_{\rm Pr}} , \qquad (1)$$

$$S_{\rm SL} = \frac{(d_{\rm Y}/\rho_{\rm Y})S_{\rm Y} + (d_{\rm Pr}/\rho_{\rm Pr})S_{\rm Pr}}{(d_{\rm Y}/\rho_{\rm Y}) + (d_{\rm Pr}/\rho_{\rm Pr})} , \qquad (2)$$

where d is the total thickness of the superlattice sample, $d_{\rm Y}$ is the total thickness of YBCO layers, and $d_{\rm Pr}$ is that of PrBCO layers. The calculated resistivity and TEP of superlattice sample A are shown in Figs. 1 and 2 by dotted lines, which are close to the behaviors of that of YBCO and are much less than the experimental results for the superlattice sample. The reason is that the conductance of the YBCO layers derived from the resistivity of the bulk sample is much higher than that of the PrBCO layers and, consequently, dominates the transport



FIG. 3. (a) Resistivity of PrBCO film. (b) Thermoelectric power of PrBCO film.

behavior. It seems that Eqs. (1) and (2) are correct in form, but the resistivity and TEP of the YBCO and PrBCO layers are different from that of bulk samples.

A. Resistivity

Usually, the total thickness of the superlattice sample is used to calculate its resistivity from the measured resistance. Since the resistivity of semiconducting PrBCO layers is much higher than that of YBCO layers, at least in low temperatures, therefore the average resistivity of the superlattice sample is higher than that of YBCO. This has been proved by Li et al., who investigated the YBCO/PrBCO superlattice with the same thickness of YBCO layers but a different thickness of PrBCO layers and found that the sheet conductance of a single YBCO layer (the measured sheet conductance divided by the number of periods) at 100 K does not change with the thickness of PrBCO lavers.² However, the calculated resistivity is still higher than that of the bulk YBCO sample when only the thickness of YBCO layers is used. First, this might due to charge transfer between the YBCO layers and PrBCO layers,^{6,8,10} which leads to a decrease of hole concentration in the YBCO and, as a result, a higher resistivity. This may be a substantial factor, but it is difficult to be estimated quantitatively.

Second, we believe the size effect should be considered. Since the mean free path of the charge carriers is about several nanometers,²⁰ which is comparable to the thickness of the YBCO layer in the superlattice, the diffusion scattering by the interface between YBCO and PrBCO layers, where there are always some structural defects⁵ may increase the resistivity. According to Cottey's model the ratio of conductance of thin film σ_f to that of bulk sample σ_b is²¹

$$\frac{\sigma_f}{\sigma_b} = C(\mu) = \frac{3}{2}\mu \left[\mu - \frac{1}{2} + (1 - \mu^2)\ln\left[1 + \frac{1}{\mu}\right]\right], \quad (3)$$

where

$$\mu = \frac{d}{\lambda_0(1-p)}, \quad 0$$

and d is the thickness of the thin film, λ_0 is the mean free path of the carrier, and p is the specularity parameter. In the case of YBCO, the mean free path can be evaluated as²²

$$\lambda_0 = v_F \tau = \frac{\hbar v_F}{2\pi \lambda k_B T} , \qquad (5)$$

where v_F is the Fermi velocity, τ is the relaxation time, and λ is the McMillan coupling parameter. The conductivity of bulk YBCO samples, σ_b^Y , can be regarded as the measured one in the YBCO film, since its thickness is much greater than the mean free path. As the conductance of the superlattice is mainly determined by the YBCO layers in low temperature, the average conductivity of YBCO layers for the superlattice sample A is roughly $\sigma_{SL}^Y = \sigma_{SL}/0.7$, where σ_{SL} is the average conductivity when the total thickness is used and the factor 0.7 comes from the fact that the thickness of YBCO layers is 70% of the total thickness for this sample. σ_{SL}^{Y} is certainly not σ_{f}^{Y} as defined in Eq. (3) because of additional charge-transfer effect. We simply assume that the difference between σ_{f}^{Y} and σ_{SL}^{Y} is proportional to σ_{b}^{Y} , i.e.,

$$\sigma_f^{\rm Y} = \sigma_{\rm SL}^{\rm Y} + q \sigma_b^{\rm Y} . \tag{6}$$

Thus, using Eq. (3), we have

$$\frac{\sigma_{\rm SL}^{\rm Y}}{\sigma_b^{\rm Y}} = \frac{\sigma_f^{\rm Y}}{\sigma_b^{\rm Y}} - q = C(\mu) - q \quad . \tag{7}$$

Taking $v_F \approx 7 \times 10^7$ cm/s, $\lambda \approx 1.0$,²⁰ d = 8.4 nm in sample A, and assuming that the coefficient q in Eqs. (6) and (7) is temperature independent, Eq. (7) fits the experimental data quite well below 200 K (Fig. 4) with q = 0.48 and $p \approx 0.4$. The value of $C(\mu)$ is about 0.81-0.92, varying with temperature. From Eq. (3), roughly speaking, about 10% of the increasing resistivity of the individual YBCO layer in superlattice samples comes from the interface scattering. The deviation in high temperature is probably due to the contribution of the PrBCO layers which has a semiconductorlike resistivity-temperature dependence with lower resistivity at high temperatures. The charge-transfer effect may enhance the reduction of PrBCO layers rises according to Eq. (1).

B. Thermoelectric power

Just like the case of resistivity, the YBCO layers may dominate the TEP behavior of the superlattice samples and the interface scattering may also affect the TEP of the YBCO layers. According to a simplified model, the difference between the TEP of thin film and that of a bulk sample is²¹

$$S_f - S_b = -\frac{\pi^2 k_B^2 T}{8eE_F} \frac{(1-p)\lambda_0}{d} \left[\frac{\partial \ln \lambda_0(E)}{\partial \ln E} \right]_{E_F} .$$
 (8)



FIG. 4. The ratio of the average conductance of a YBCO layer in the superlattice sample A, σ_{SL}^{Y} , to the conductance of bulk sample σ_{b}^{Y} (triangle) and the fitting curve (solid line).

Since the mean free path λ_0 is proportional to 1/T according to Eq. (5) and generally the above differential term is considered to be temperature independent, this effect has no influence on the slope of S(T) and only adds a constant to the TEP. With an assumption that $\lambda_0(E) = v_F / \tau \propto E^{1/2}$ and taking $p \approx 0.4$, $E_F = 0.1 \text{ eV}$,¹⁰ we found that the change is not greater than $1 \mu \text{V/K}$, which is very small compared to the difference between the TEP of YBCO and that of the superlattice and can be neglected.

In view of several studies, 12-14,23 the TEP of YBCO is very sensitive to the oxygen content. For oxygen rich samples ($\delta < 0.1$), the TEP is negative and increases with increasing temperature, i.e., dS(T)/dT > 0, in the range from 100 to 300 K. For the samples with δ just above 0.1, the TEP has small positive value (<5 μ V/K) and weak temperature dependence. For higher δ values $(0.2 < \delta < 0.5)$, the TEP becomes large (~10 μ V/K) with an obvious negative slope dS(T)/dT. Cohn et al. have measured the TEP of untwinned YBCO single crystals.²⁴ They found that the *a*-axis TEP is quite similar to the inplane TEP of other cuprates, such as the Bi and Tl systems,²⁵ which have negative slope dS(T)/dT, and the chain TEP, which is strongly oxygen dependent, increases with increasing temperature and saturates in high temperature.

Low concentration of carriers usually leads to a large TEP value.^{23,26} Sometimes, the TEP of the high- T_c superconductor in the *ab* plane can be written in the form $S(T) = \alpha + \beta T$.²⁷ The constant term α may be attributed to the electron-phonon enhancement effect²⁸ or to the contribution of spin entropy.²⁹ The two models all suggest a large value of α for low concentration of carriers. Thus we speculate that the large positive value of the TEP (10-20 μ V/K) in our superlattice samples might result from the reduction of the hole concentration in the YBCO layers, which is consistent with the charge transfer model.

The most striking feature of the TEP in our superlattice samples is the positive slope dS(T)/dT, while the TEP has large positive value. This TEP behavior differs from that of YBCO with different oxygen content, negative TEP value with positive slope dS(T)/dT, or positive value with negative value. It also differs from that of YPrBCO as well as the oxide superconductors of the Bi and Tl systems, which usually show negative slope.²⁵ At present, we do not quite understand it. A possible explanation is that it results from contribution of Cu-O chains. Even though charge transfer leads to a reduction of carrier density and makes the TEP to be positive, the chain structure remains perfect because the oxygen in YBCO layers has not been taken out. So the contribution to TEP from chains is still important in YBCO layers. It results in a positive slope dS(T)/dT as that in oxygenrich samples. The PrBCO layers, which have a large positive value of S and a positive temperature coefficient of S(T), might also make some contribution to the increasing tendency of the TEP in the superlattice samples, since it is possible that the conductivity in the PrBCO layers is improved due to charge transfer. On the other hand, the increase of hole concentration may lead to a decrease of



FIG. 5. The phonon-drag thermoelectric power S_g vs 1/T for the superlattice sample A and YBCO film. The lines are guides to the eye.

the TEP values.¹⁹ These two effects may cancel with each other according to Eq. (2). In short, the contribution from PrBCO layers is hard to estimate.

There is a broad peak just above T_c in the S(T) curves of the superlattice sample A and YBCO film (Fig. 2). The peak in the TEP of YBCO is considered to come from the phonon-drag effects.³⁰ We believe that the peak in TEP of YBCO/PrBCO has the same origin. If the linear part is subtracted from the total TEP, the phonondrag TEP, S_{g} , which is inversely proportional to temperature, can be obtained. According to a simple theory,¹⁹ the slope of S_g vs 1/T is inversely proportional to the concentration of carriers. From Fig. 5, it can be seen that the slope of the superlattice is greater than that of YBCO, which means that hole concentration in the YBCO layers of the superlattice is lower. This is reasonable. Since the intensity of the phonon drag is related to the structure of the lattice, the more perfect the lattice structure is, the more obvious the phonon drag. The YBCO layers in the superlattice sample B is thinner than that of sample A and the defects in the interface are more important, so the phonon-drag peak is not obvious in the S(T) curve of sample **B**.

IV. CONCLUSION

In summary, we have measured the resistivity and the TEP of two YBCO/PrBCO superlattice samples in the temperature range 80-270 K. It was found that the resistivity is mainly determined by the YBCO layers, which has higher resistivity than that of bulk YBCO film. The contribution of the interface scattering can be described by Cottey's model. At temperatures above 200 K, the influence of the PrBCO layers appears. The values of TEP of the superlattice are greater than that of the

slightly oxygen-deficient YBCO sample and the slope of the linear part of S(T) changes to positive, which is also different from the typical negative slope observed in the in-plane TEP of the Bi and Tl systems. The broad peak in the S(T) curve of the superlattice sample A just above T_c may be attributed to the phonon-drag effect.

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