High-temperature resistivity of RBa$_2$Cu$_3$O$_{7-y}$, where R = La, Pr, Nd, Sm, Eu, Dy, Ho, Er, and Tm

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The electrical resistivity ρ of RBa$_2$Cu$_3$O$_{7-y}$, where R = La, Pr, Nd, Eu, Dy, Ho, Er, Sm, and Tm has been measured from room temperature up to 900 K. No essential difference in the transport properties was found except for LaBa$_2$Cu$_3$O$_{7-y}$ and PrBa$_2$Cu$_3$O$_{7-y}$ which shows an anomaly in the electrical resistivity. SmBa$_2$Cu$_3$O$_{7-y}$ samples with various oxygen contents are studied in detail. Metallic behavior and linear temperature dependence of ρ have been found for SmBa$_2$Cu$_3$O$_{7-y}$ with $0 < y < 0.227$. The linear temperature dependence of ρ implies weak electron-phonon coupling, which suggests that other mechanisms for high-T_c superconductivity are needed.

There has been much work[1,2] on the superconducting properties of RBa$_2$Cu$_3$O$_{7-y}$. The x-ray data show that there is an orthorhombic-to-tetragonal structural transformation.[3] RBa$_2$Cu$_3$O$_{7-y}$, where R is a rare-earth element, shows similar electric and magnetic properties.[4,5] In many reports it has been shown that an oxygen content of 7.0 gives the best orthorhombic superconducting phase of YBa$_2$Cu$_3$O$_{7-y}$. The resistivity is strongly related to the oxygen deficiency at high temperatures.[6] Hence, the oxygen content has been shown to be one of the main factors which affects the physical properties of the high-T_c superconductor YBa$_2$Cu$_3$O$_{7-y}$.[7-13] In this work we will study the electrical resistivity of RBa$_2$Cu$_3$O$_{7-y}$, where R = La, Pr, Nd, Eu, Dy, Ho, Er, Sm, and Tm. SmBa$_2$Cu$_3$O$_{7-y}$ was chosen as a prototype compound in studying the effects of oxygen contents on the metal-semiconductor transition and superconductivity of RBa$_2$Cu$_3$O$_{7-y}$. We derive the electron-phonon coupling constant λ_{tr} from the electric and magnetic data. The implication of its value will be discussed.

The sample preparation used in this study was reported previously.[4] The high- and low-temperature resistivity (ρ) measurements were performed on two separate four-probe instruments with automated data acquisition. At 300 K and higher temperatures, four Pt leads were screwed tightly to a rectangular sample; silver paint was applied to attach four Cu wires to the sample for low-temperature resistivity experiments. A well-calibrated Pt thermometer was used to indicate the temperature in the high-temperature range, and a reliable, calibrated silicon diode was the temperature probe in the low-temperature range. For both high- and low-temperature regions, the resistivity was obtained from Linear Research LR 400 resistance bridge. Data points were less than 0.2 K apart. These ρ-versus-temperature data were then used directly in calculating derivatives and in other analyses. To prevent the reabsorption of oxygen during high-temperature resistivity measurements, about 1 atm flowing He gas was applied to the chamber. After heating the sample to a certain maximum heating temperature T_m, we cooled and heated the sample several times with the highest temperature not exceeding T_m and found that the resistivity curves coincide. This tells us that the oxygen content of the sample in the cooling resistivity measurements remained the same as that at T_m in our experiments.

Thermogravimetric analysis (TGA) data were taken on a du Pont 9900 thermal analysis system also with 1 atm flowing He gas in order to indicate the oxygen content for the sample at T_m during high-temperature resistivity measurements.

Figure 1 shows lnρ as a function of T for RBa$_2$Cu$_3$O$_{7-y}$. The arrows indicate the temperatures where the structure transformation occurs for RBa$_2$Cu$_3$O$_{7-y}$. The rapid increase in ρ at $T > 500$ K is due to the desorption of oxygen. Resistivity on cooling differed from

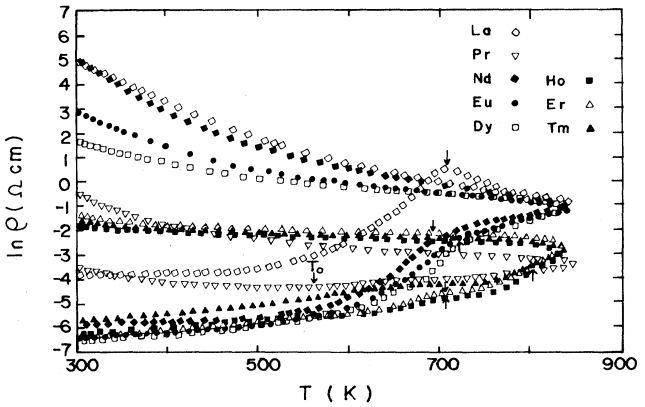

FIG. 1. Temperature dependence of lnρ for RBa$_2$Cu$_3$O$_{7-y}$, where R = La, Pr, Nd, Eu, Dy, Ho, Er, and Tm. The arrows indicate the temperature where the structural transformation occurs. Above T_0, the resistivity increases for sample PrBa$_2$Cu$_3$O$_{7-y}$.

that on warming due to the reduction in oxygen content. This is different from previous work[8] performed in air or oxygen which did not show hysteresis if the cooling was swept slowly enough to allow the oxygen to reach equilibrium. Similar features were found in the temperature dependence of ρ, except for the cases of $LaBa_2Cu_3O_{7-y}$ and $PrBa_2Cu_3O_{7-y}$. For $LaBa_2Cu_3O_{7-y}$ the temperature dependence of ρ has a relative maximum at 700 K, and a concave upward feature after cooling the sample from high temperature. This anomaly in the resistivity maximum has been rechecked and indeed exists. Two main factors can be the causes: (1) a structural transformation which changes ρ from metallic to semiconductive behavior, and (2) inhomogeneity in samples and thermodynamic reaction. While this is less likely, it cannot completely be ruled out. For $PrBa_2Cu_3O_{7-y}$ the sample shows semiconducting behavior. As the temperature of the sample is increased from room temperature, the resistivity decreases. As one increases the temperature above T_0 (as shown in Fig. 1), the resistivity increases. In this range ($T > T_0$) the desorption of oxygen causes the electrical resistivity to increase.

Figure 2 shows the wt.% of $SmBa_2Cu_3O_{7-y}$ as a function of temperature. Since Sm, Ba, and Cu are unlikely to come out of bulk materials, it is assumed that the weight losses are due to the oxygen only. Therefore, we may calculate the corresponding $7-y$ values, plotted on the right-hand side of Fig. 1, according to the following relation:

$$\frac{\text{wt.\%}}{100} = \frac{\text{weight of } SmBa_2Cu_3 + 15.996(7-y)}{\text{weight of } SmBa_2Cu_3O_7} . \quad (1)$$

In this figure, we find that a rapid increase in the oxygen desorption rate occurs at $T > 730$ K and the data suggest that there is a orthorhombic-to-tetragonal structural transformation. This observation will also be discussed in relation to the resistivity data. The results in Fig. 2 were our calibration standard in determining the oxygen content for $SmBa_2Cu_3O_{7-y}$ heated to T_m. It is reasonable to conclude that, because we have done the resistivity measurement along the heating and cooling curves several times after T_m annealing and found that the cooling and heating curves overlap together, the phase is stabilized after T_m and oxygen desorption or reabsorption does not occur under flowing He atmosphere.

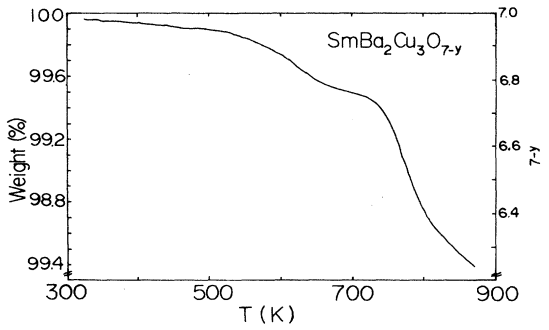

FIG. 2. Temperature dependence of weight and oxygen content in $SmBa_2Cu_3O_{7-y}$.

For high-temperature resistivity measurements, $RBa_2Cu_3O_{7-y}$ was cut into rectangular pieces. The precise relations between y and T_m for typical $SmBa_2Cu_3O_{7-y}$ samples are listed in Table I (the symbols are the same for Figs. 3 and 6).

The temperature dependence of ρ in high-T_c oxides has been studied for $YBa_2Cu_3O_{7-y}$ up to 1200 K.[14] However, in these studies the thermal desorption of oxygen at high temperature changed the oxygen contents and the temperature dependence of ρ, which depends mainly on the phonon scattering and vacancy scattering. If one can fix the oxygen content in some way, then the temperature dependence of ρ depends only on the phonon scattering. To obtain a sample with fixed oxygen content and also perform the resistivity measurement at high temperature ($T < T_m$), we have annealed the sample in helium atmosphere and simultaneously monitored the electrical resistivity. The basic idea is that if we heat the sample up to the highest temperature T_m (heating rate $\sim 5°$/min), then only a finite amount of oxygen will remain in the sample. If the sample does not absorb oxygen while it is cooled from T_m then the oxygen content is fixed after cooling from T_m. Indeed this idea has been proven to be correct in the resistivity measurement, as we mentioned earlier.

Figure 3 shows the resistivity loops for samples which show superconductivity after cooling from T_m. Several interesting features were found in these loops. (1) For samples with $y < 0.227$ (see Table I), the temperature dependence of ρ in the cooling curve from 650 K down to T_c is linear. (2) For samples with $0.266 < y < 0.446$, the temperature dependence of ρ in the cooling curve shows semiconductivity before the superconducting transition. And (3), hysteresis loops were found for all samples. The linear temperature dependence of ρ up to high temperature for fixed y is the essential finding in this work, especially the temperature dependence of ρ up to T_m for sample E which has the highest T_m and still shows metallic behavior.

TABLE I. Superconducting transition temperatures T_c (50%), y values, and maximum heating temperatures T_m, during high-temperature resistivity measurements in $SmBa_2Cu_3O_{7-y}$.

Samples	Symbols	T_c (50%) (K)	T_m (K)	y
A		95.5		0
B	○	90.7	521	0.059
C	▲	88.1	535	0.073
D	□	87.0	566	0.100
E	▼	91.1	584	0.122
F	◇	83.6	623	0.187
G	●	87.8	659	0.227
H	▽	67.6	703	0.266
I	·	59.8	717	0.291
J	×	44.7	738	0.446
K	◆	· · ·	752	0.533
L	△	· · ·	755	0.564
M	■	· · ·	778	0.657
N	+	· · ·	809	0.735

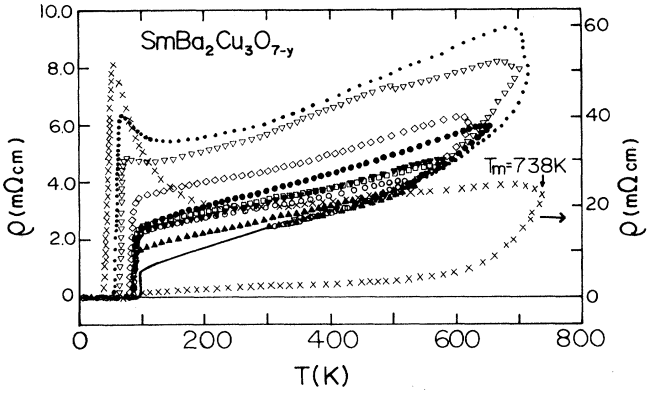

FIG. 3. Resistivity vs temperature for samples B–J, which are superconducting as cooled from T_m. The symbols used here are the same as those shown in Table I. T_m for sample J is marked.

From the results of the samples which show metallic behavior, we predict that a linear dependence of ρ for metallic RBa$_2$Cu$_3$O$_{7-y}$ samples should extend even to higher temperature ($T > 650$ K) if the oxygen content ($y \sim 0$) can be stabilized.

Figure 4 shows the room-temperature resistivity and the superconducting transition (50% resistive transition) as a function of oxygen content $7-y$. The room-temperature resistivity rises sharply for $y > 0.5$. Samples with $y > 0.5$ are semiconducting while samples with $y < 0.5$ show superconducting behavior.

In this work we have used a heating cycle to desorb oxygen from the sample. If the heating rate is too fast, it is likely that the resulting oxygen content is nonuniform. That is, the cores of the grains would have a larger oxygen content and have a higher T_c. To see whether the samples

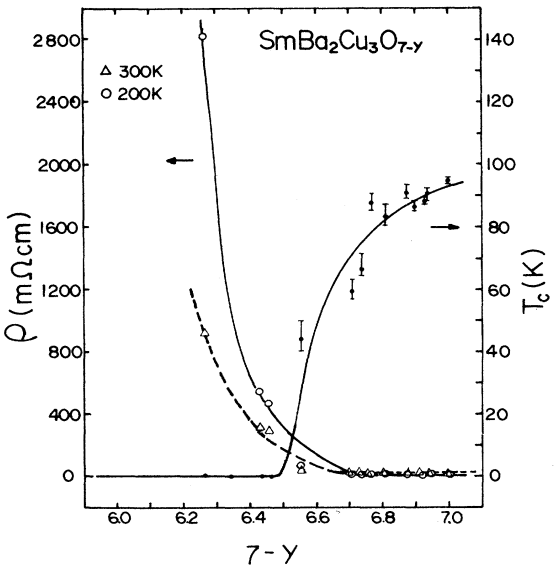

FIG. 4. Room-temperature resistivity and superconducting transition temperature (50% resistive transition) as a function of $7-y$.

are uniform or not, magnetic susceptibility, which is a probe of the bulk superconductivity, is measured. The magnetic susceptibility shows an onset superconducting transition temperature comparable to the resistive transition, indicating the samples are uniform.

Figure 5 shows $d\rho/dT$ as a function of T for Sm-Ba$_2$Cu$_3$O$_{7-y}$. The resistivity is essentially linear at low temperature. At higher temperature one finds a thermally activated departure from the linear law by oxygen desorption.[14,15] The break-in slope at high temperature corresponds closely with the temperature $T_{OT,\rho}$ of the orthorhombic-to-tetragonal (OT) structural transformation which is at about 730 K (minimum in $d\rho/dT$).

The orthorhombic-to-tetragonal phase transformation has been observed in YBa$_2$Cu$_3$O$_{7-y}$ under different oxygen partial pressures. Detailed temperature-dependent neutron-diffraction measurements in atmosphere with various oxygen partial pressures reveal a rather complicated relation between temperature, oxygen deficiency, and the extent of the tetragonal-to-orthorhombic distortion. Apparently the transformation is induced through a disordering of the O vacancies as the temperature increased.

In Table II, the $T_{OT,\rho}$'s derived from the high-temperature resistivity data are shown for RBa$_2$Cu$_3$O$_{7-y}$. We note that the temperature at which the phase transformation occurs depends on the oxygen partial pressure in the ambient atmosphere which, in our experiments, is helium. So the derived transformation temperature should be lower compared with results obtained under higher oxygen partial pressures.[16]

Figure 6 shows $\ln\rho$ vs $1/T$ in the high-temperature semiconductive range for SmBa$_2$Cu$_3$O$_{7-y}$. In this range the temperature dependence of ρ follows the relation:

$$\rho = \rho_0 \exp(E/k_B T) , \tag{2}$$

where E is the activation energy. If one fits the data to Eq. (2), we get the activation energy E as shown in the inset of Fig. 6. The activation energies for SmBa$_2$Cu$_3$O$_{7-y}$ vary from about 50 to about 90 meV as y varies from 0.55 to 0.72. A minimum in activation energies was found.

In Table II the activation energies for RBa$_2$Cu$_3$O$_{7-y}$ are shown for comparison. The E value in Table II for Eu

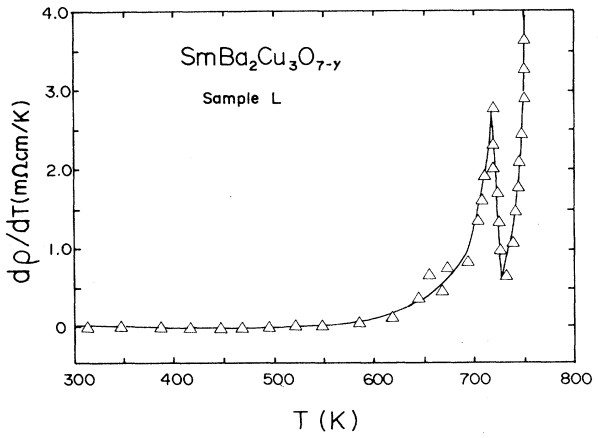

FIG. 5. $d\rho/dT$ as a function of T for SmBa$_2$Cu$_3$O$_{7-y}$.

TABLE II. The structural transformation temperature (T_{OT}) derived from resistivity measurement; $7-y$ is the oxygen content after heating the sample to the highest temperature, E and ρ_0 are obtained from Eq. (2).

RBa$_2$Cu$_3$O$_{7-y}$ (R=)	T_{OT}, from resistivity (K)	y	E (eV)	ρ_0 (Ω cm)
La	712	0.57	0.213	0.0830
Nd	694	0.77	0.243	0.0157
Eu	723	0.85	0.153	0.0476
Dy	705	0.83	0.100	0.0194
Ho	710	...	0.025	0.0596
Er	708	0.86	0.037	0.0593
Tm	700	0.73	0.038	0.0702

and Dy compounds, which have oxygen deficiencies of about 0.85 and 0.83, respectively, is of the same order of magnitude as that extrapolated from the Sm compound. For La and Nd compounds the activation energy is a little bit larger. We attribute the larger value of E to a small amount of semiconducting or other phases present in the samples. This can be seen from the relative low T_c's for La and Nd compounds (Fig. 7), which suggest that La and Nd compounds prepared under present conditions are not as good in their metallic properties as that of other RBa$_2$Cu$_3$O$_{7-y}$ samples reported here.

Samples exhibiting semiconductivity on cooling are not superconducting above 4.2 K. It has been reported[16] that superconductivity exists only for $y \leq y_c \sim 0.5$–0.6, where y_c is the critical value above which no superconductivity was found. For our samples, the $y > y_c$, no superconductivity was found.

For the magnetic measurement the samples were ground into fine powder and sieved through 400 mesh. The size of the particles, about 5 μm,[10] was determined using a microscope with a standard calibration. The magnetic susceptibility was calibrated with Nb powder with a size of about 80 μm. No correction in magnetization was used in our calculation because we used Nb powders for calibration.

Figure 7 shows the temperature dependence of the ac susceptibility for RBa$_2$Cu$_3$O$_{7-y}$. The data derived from

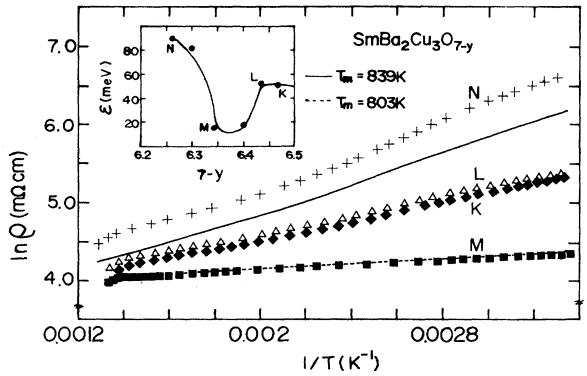

FIG. 6. lnρ vs $1/T$ for semiconductive samples.

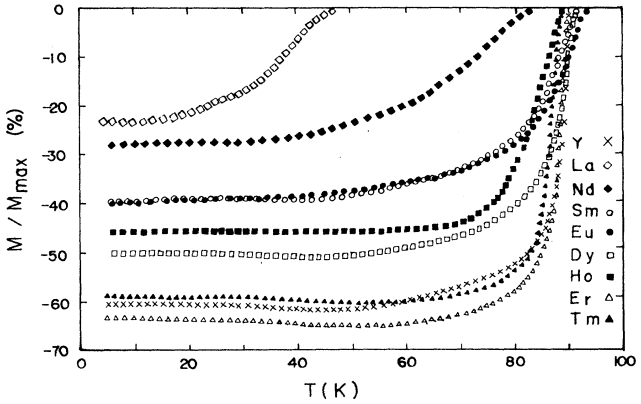

FIG. 7. Temperature dependence of M/M_{max} for RBa$_2$Cu$_3$O$_{7-y}$, where R=Y, La, Nd, Sm, Eu, Dy, Ho, Er, and Tm.

the magnetic measurements are shown in Table III. For a superconducting sphere, the relation between M/M_{max} and $x=\lambda_L/a$ has been reported in Ref. 17, i.e.,

$$M/M_{max}=1-3x\coth(1/x)+3x^2, \tag{3}$$

where a is the average diameter of the particle. Some authors[18,19] have used Eq. (3) to study the superconducting oxides. If we assume a temperature dependence of $\lambda_L(T)$ given by

$$\lambda_L(T)=\lambda_L(0)[1-(T/T_c)^4]^{-1/2}, \tag{4}$$

where $\lambda_L(0)$ is the London penetration depth at zero temperature, Eqs. (3) and (4) can be used to derive $\lambda_L(T)/\lambda_L(0)$ from the measured M/M_{max}.

Figure 8 shows the temperature dependence of $\lambda_L(T)/\lambda_L(6\text{ K})$ for RBa$_2$Cu$_3$O$_{7-y}$ samples, where R is Sm and Er. Different features in $\lambda_L(T)/\lambda_L(6\text{ K})$ were found. In our original planning of the experiment we hoped to see a consistent behavior in $\lambda_L(T)/\lambda_L(6\text{ K})$ because similar electric and magnetic properties have been

TABLE III. X-ray density is derived from the x-ray lattice parameters (taken from Ref. 4), and $T_{c,onset}$ is the onset superconducting transition temperature. M/M_{max} is the relative magnetization. $\lambda_L(\sim 6\text{ K})$ is the London penetration depth and λ_{tr} is the electron-phonon coupling constant for RBa$_2$Cu$_3$O$_{7-y}$.

RBa$_2$Cu$_3$O$_{7-y}$ (R=)	X-ray density (g/cm^3)	$T_{c,onset}$ (K)	M/M_{max} (%)	$\lambda_L(\sim 6\text{ K})$ (μm)	λ_{tr}
Y	6.324	90.53	60.45	0.391	0.275
La	6.577	47.7	23.91	1.078	0.212
Nd	6.746	87.6	28.01	0.802	0.113
Sm	6.853	92.3	40.25	0.683	0.139
Eu	6.880	93.8	39.91	0.691	0.145
Dy	7.057	91.19	50.01	0.528	0.200
Ho	7.095	89.39	45.68	0.593	0.257
Er	7.139	91.10	62.70	0.356	0.375
Tm	7.186	90.08	58.99	0.409	0.371

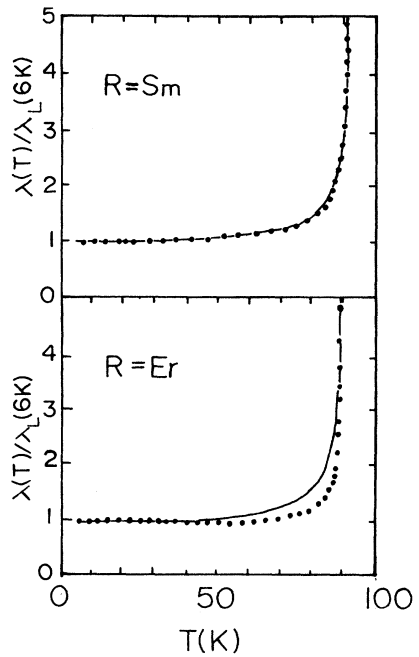

FIG. 8. $\lambda_L(T)/\lambda_L$ (6 K) as a function of temperature for RBa$_2$Cu$_3$O$_{7-y}$, where R =Sm and Er.

reported.[4] However, the results obtained here are not quite similar. Perhaps they are due to (1) different grain structures or (2) different size distribution.

The magnetic field penetration depth can be derived if we know the average size of the powders. Using an average value about 5 μm, which is estimated using an optical microscope, the derived $\lambda_L(0)$'s are shown in Table III, which has the same order of magnitude as that reported in Ref. 13. However the $\lambda_L(0)$ is a bit larger compared with that derived from the muon-spin resonance.[20] Because of the anisotropy of the field penetration depth, the penetration depth along the a-b plane is believed to be larger than that along the c axis. Perhaps the muon-spin resonance measured the smaller value of the anisotropy while the magnetic method measured the larger value of anisotropy.[19]

Chakraborty, Pickett, and Allen[21] have proposed a scheme for analyzing the phonon-limited resistivity of metals. The resistivity can be written as

$$\rho=[(n/m^*)e^2]^{-1}(1/\tau_{e\text{-}p}) , \tag{5}$$

$$n/m^* = \tfrac{1}{3}N(0)\langle v^2\rangle , \tag{6}$$

where $1/\tau_{e\text{-}p}$ is the electron-phonon scattering rate, m^* is the effective electron mass, n is the electron density, e is the electronic charge, $N(0)$ is the density of states at Fermi surface, and $\langle v^2\rangle$ is the mean value of v^2. A simple formula for the resistivity which holds for $T>\Theta_D$ is

$$\rho=[(n/m^*)e^2]^{-1}\hbar^{-1}(2\pi\lambda_{\rm tr}k_BT) . \tag{7}$$

In the free-electron model, λ_L can be expressed as

$$\lambda_L^2=m^*/(ne^2\mu_0) , \tag{8}$$

where μ_0 is the permeability in free space.

Putting $\lambda_L(0)$ into Eq. (8), we obtain m^*/n. Using these values of m^*/n and Eq. (7), we derive $\lambda_{\rm tr}$'s for RBa$_2$Cu$_3$O$_{7-y}$ as shown in Table II. We noted that the ρ vs T data should be linear for $T>\Theta_D$ in order to use Eq. (7) to derive $\lambda_{\rm tr}$'s. Indeed, we have found a linear temperature dependence of $\rho(T)$ up to 600 K for RBa$_2$Cu$_3$O$_{7-y}$ in the metallic region. There is a scatter in the derived $\lambda_{\rm tr}$'s, which are perhaps due to different grain structures. It should be noted that we have overestimated the value of λ_L, probably at least a factor of 2, due to the uncertainty in $\lambda_L(0)$ determined from the magnetic measurement, which is about four times larger than that derived from the muon-spin resonance.[20] In addition, we believe there are anisotropy effects in $\lambda_L(0)$ which are not considered in our estimate of $\lambda_L(0)$.

Because the coupling constant in the theory of superconductivity is closely related to the coupling constant $\lambda_{\rm tr}$ from resistivity,[21] a study of $\lambda_{\rm tr}$'s will help us understand the superconductivity of the oxides. Recent calculations reported in Ref. 22 show that $\lambda=0.31$ for YBa$_2$Cu$_3$O$_{7-y}$. This is very similar in magnitude to the obtained $\lambda_{\rm tr}$'s of RBa$_2$Cu$_3$O$_{7-y}$. This is not surprising because theory[21] gives closely related expressions for these numbers. Our present results show weak electron-phonon interaction, because the coupling constant would predict $T_c\sim 0$ K if McMillian's[23] theory is used with $\mu^*=0.1$ and if we assume $\lambda=\lambda_{\rm tr}$.

In conclusion, we have systematically measured the temperature dependence of ρ up to 900 K for RBa$_2$Cu$_3$O$_{7-y}$ samples. A linear temperature dependence of ρ was found and the ρ-vs-T curve in a cycle is hysteretic. The hysteresis in ρ is reasonable because it was performed under helium atmosphere, so the oxygen content is different after heating the sample up to high temperature. The coupling constant $\lambda_{\rm tr}$, extracted from the linearity and field penetration depth, shows weak electron-phonon coupling. This suggests high-T_c superconductivity must be nonphononic in origin.[15]

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