Thermopower of $RBa_2Cu_3O_{7-x}$ (R=Y,Er)

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Resistance and absolute thermopower of high-T_c oxide superconductors $RBa_2Cu_3O_{7-x}$ (R=Y,Er) synthesized by a hot-press sintering technique have been measured in the temperature range of 4–300 K. Resistive behavior of all samples is found to be metallic. The thermopower for both compositions is positive above the superconducting transition temperature. The thermopower of *as-prepared* $ErBa_2Cu_3O_{7-x}$ samples shows a weak temperature dependence with a negative slope near room temperature, which increases in magnitude as T_c is approached from above. No strong thermopower peak just above T_c has been observed either in $YBa_2Cu_3O_{7-x}$ or $ErBa_2Cu_3O_{7-x}$. Reannealing of an $ErBa_2Cu_3O_{7-x}$ sample at 500 °C for 12 h in flowing oxygen increases the magnitude of the thermoelectric power and the magnitude of its slope without affecting T_c. Comparison of the results with available theory indicates that there is no completely satisfactory theory of electrical transport in these superconducting oxides.

INTRODUCTION

Among the family of high-T_c oxide superconductors, having chemical composition $RBa_2Cu_3O_{7-x}$, where R is a rare-earth metal, $YBa_2Cu_3O_{7-x}$ has been investigated extensively.[1] Only a few studies on other rare-earth-based oxide superconductors are available in the literature. Normal-state electron-transport properties, namely, resistivity ρ, Hall coefficient R_H, and thermoelectric power S of polycrystalline as well as single crystals of $YBa_2Cu_3O_{7-x}$ have been investigated extensively to obtain clues to the possible interaction responsible for the high superconducting transition temperature of these 1:2:3 superconductors. Most reported measurements of ρ, R_H, and S on polycrystalline and single-crystal $YBa_2Cu_3O_{7-x}$ superconductors show that the sign of majority carriers is positive.

The temperature dependence of these transport coefficients in $YBa_2Cu_3O_{7-x}$ as well as other $RBa_2Cu_3O_{7-x}$ superconductors is observed to be quite different than expected from a simple metallic conduction mechanism. For example, R_H increases as T_c is approached from room temperature;[2] the temperature coefficient of resistance is positive or negative depending upon the oxygen content in the sample, which also determines the magnitude of the thermopower.[3–7] A strong peak in S just above T_c was reported for some $YBa_2Cu_3O_{7-x}$ samples.[8–11] Wide variations of measured transport coefficients of $YBa_2Cu_3O_{7-x}$ samples have been attributed to different methods of sample processing, oxygen deficiency, and granular structure of polycrystalline samples. However, recently a number of results have been reported on single crystals of $YBa_2Cu_3O_{7-x}$, and surprisingly there seems to be relatively more disagreement among these results, not only of the magnitudes but also in the temperature dependence of electron transport coefficients, than those reported for polycrystalline samples.

While a number of $RBa_2Cu_3O_{7-x}$ samples[1,5,12] exhibit superconductivity with a similar T_c and have a similar structure, there have been few attempts to study transport properties of materials other than $YBa_2Cu_3O_{7-x}$. Recently, a detailed study on $EuBa_2Cu_3O_{7-x}$ has been reported.[4] Besides this study, there are scanty reports on other $RBa_2Cu_3O_{7-x}$ systems. It is therefore useful to study other such systems to gain more insight into 1:2:3: superconductors.

In this report we present results of resistance and thermopower measurements on $ErBa_2Cu_3O_{7-x}$ and $YBa_2Cu_3O_{7-x}$ superconductors synthesized by a hot-press sintering technique.[13] $RBa_2Cu_3O_{7-x}$ superconductors prepared this way have been shown to have higher densities, to have a sharper and hysteresis free resistive transition and to be able to carry larger current densities.[13] Our main interest was in $ErBa_2Cu_3O_{7-x}$, but an $YBa_2Cu_3O_{7-x}$ superconductor was also studied for comparison with the results of others to determine the influence of the hot-press sintering process on the thermopower.

EXPERIMENTAL DETAILS

Oxide superconductors $YBa_2Cu_3O_{7-x}$ and $ErBa_2Cu_3O_{7-x}$ were prepared by a hot-press sintering technique described in detail elsewhere.[13] Rectangular samples of approximately 1- to 1.5-mm thickness, 5-mm long, and 2-mm wide were cut from the hot-pressed cylinders. They were single phase with a density of 5.8 g/cm^3 and 5.0 g/cm^3 for Er and Y samples, respectively (i.e., about 80% of the theoretical densities). Two $ErBa_2Cu_3O_{7-x}$ samples, designated No. 1 and No. 2, were prepared at different times but under identical conditions.

Resistance and thermoelectric power were measured as a function of temperature on an apparatus which is described in more detail elsewhere.[14,15] Electrical contacts were made using well annealed gold foils of approximately 100-μm thickness in the following way. Two rectangular pieces of gold foil having the same width as the sample were cut, lightly sanded, thoroughly cleaned, and finally bent into an "L" shape. Two ends of the sample were lightly sanded and thoroughly cleaned with a camel hair brush before connecting them to the L-shaped gold foils using a thin layer of silver epoxy. The epoxy was cured in air at 50–75 °C for a few hours. Similar, but much narrower (width approximately 0.1–0.2 mm) gold foils were used to establish two contacts on the longer face of the sample for resistance measurements. The resistance was measured by a standard four terminal DC method in the temperature range 4–300 K. The temperature was measured with a calibrated platinum thermometer or a AuFe chromel thermocouple previously calibrated against it.

Thermopower was measured using a standard differential technique in which a well annealed, high-purity lead foil was used as reference. A sample with two gold foil contacts on the sides was mounted between two large copper blocks such that it fitted snugly between them, i.e., there was almost no spare length of gold foil between the copper block and the contact on the sample. This precaution, in addition to a wait of about 20–30 minutes after the *stabilization* of temperature of each copper block, assured nearly zero temperature gradient between a copper block and the nearest sample face attached to it. A carefully annealed 99.9999% Pb foil reference electrode was also mounted between the copper blocks in electrical contact with one end of the sample. Both copper blocks were stabilized at the same temperature, and the emf between the copper leads to the sample-Pb reference foil thermocouple was accurately measured to determine any parasitic thermal emfs in the leads. One of the blocks was heated to produce a small temperature difference ΔT (typically 0.1–0.2 K) between the blocks while the other block was maintained at the original temperature. Since these materials are poor thermal conductors it was important to wait the 20–30 minutes to stabilize the temperature gradient before measuring the thermal emf δV. The temperature of the sample and the temperature difference δT across the sample was measured using a calibrated AuFe-chromel thermocouple. The thermal emf was measured with a Keithley model 148 nanovoltmeter with its analog output connected to a digital volmeter for higher resolution. For each measurement, δT and δV were averaged over 100 readings taken at an interval of 0.5 s to reduce the error due to noise. The parasitic emf's which were quite small compared to δV were subtracted from it. The estimated absolute error in measurement of S is about 5%. Further details including experimental procedure and tests of the precision and accuracy can be found elsewhere.[14,15]

The oxygen content of the samples was not independently determined. One of the $ErBa_2Cu_3O_{7-x}$ samples was reannealed to 500 °C for 12 h in flowing oxygen after measurements were completed on the *as prepared* sample. Gold contacts and silver epoxy were carefully removed from the sample, and these contact faces were sanded to remove any epoxy contamination before reannealing. It was hoped that this reannealing would reduce the oxygen deficiency in the sample. However, we shall argue later that this procedure of reannealing most probably increased the oxygen deficiency.

RESULTS

Figures 1–3 show the temperature dependence of the resistance ratio, $r(T)=R(T)/R(300)$, and the thermoelectric power of $YBa_2Cu_3O_{7-x}$ and $ErBa_2Cu_3O_{7-x}$ (Nos. 1 and 2). All three samples show metallic behavior of the resistance, i.e., dR/dT is positive. The resistance ratio decreases linearly as the temperature is lowered until about 120 K, below which one observes a rounding of the resistive curve before becoming superconducting. The superconducting transition temperatures, defined by the intersection of a line drawn through the sharp drop of $r(T)$ and the temperature axis, are approximately 92 K and 90 K for $YBa_2Cu_3O_{7-x}$ and $ErBa_2Cu_3O_{7-x}$ samples, respectively. The midpoint temperature for the transition for $YBa_2Cu_3O_{7-x}$ is 93 K. These values agree well with those reported in the literature.[5,12,13] A straight line fit to data above 120 K gives values for $r(0)$ of 0.305, 0.281, and 0.249 and values for $dr(T)/dT$ of 0.0025 K^{-1}, 0.0024 K^{-1}, and 0.0025 K^{-1} for $YBa_2Cu_3O_{7-x}$ and $ErBa_2Cu_3O_{7-x}$ (Nos. 1 and 2), respectively. Here $r(0)$ is the extrapolated value of $r(T)$ at $T=0$. These values of $r(0)$ are comparable to or better than values reported in the literature, even for single crystals of $YBa_2Cu_3O_{7-x}$. From these values it seems that $ErBa_2Cu_3O_{7-x}$ (No. 2) is slightly more metallic than $ErBa_2Cu_3O_{7-x}$ (No. 1) or the $YBa_2Cu_3O_{7-x}$ sample, i.e., $ErBa_2Cu_3O_{7-x}$ (No. 2) has smaller scattering by grain boundaries and other defects than the other two samples. The slopes $dr(T)/dT$ are approximately the same for all samples. It has been es-

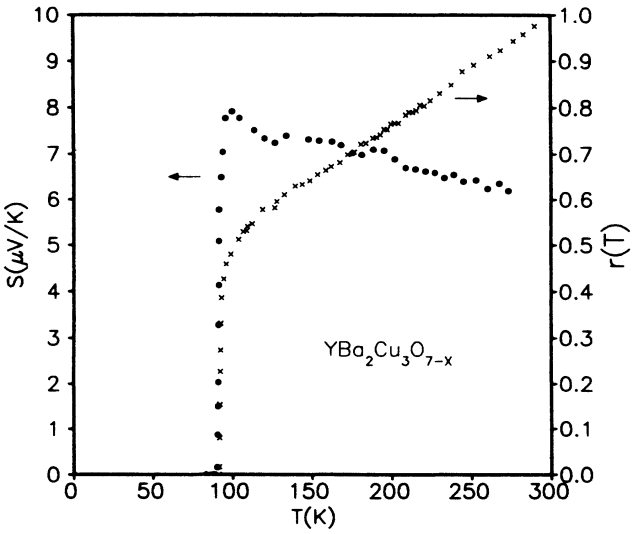

FIG. 1. Temperature dependence of the thermopower $S(T)$ and the resistance ratio, $r(T)=R(T)/R(300)$ of $YBa_2Cu_3O_{7-x}$.

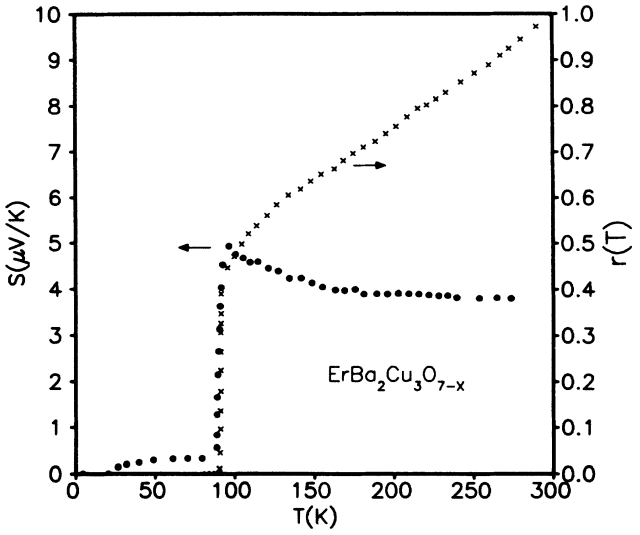

FIG. 2. Temperature dependence of the thermopower and the resistance ratio of $ErBa_2Cu_3O_{7-x}$ (No. 1).

tablished by many researchers that the content of oxygen, x, affects $r(T)$ as well as the other transport properties.[1,3,4] As x increases from zero, $r(T)$ shows a gradual transition from a metallic to semiconducting behavior. Based on these results, it seems that the x value in our samples is close to zero, but $ErBa_2Cu_3O_{7-x}$ (No. 1) is more oxygen deficient than $ErBa_2Cu_3O_{7-x}$ (No. 2) if we assume that no other factor is responsible for the different $r(0)$ and dr/dT values for these two samples prepared under identical conditions. Values of room temperature resistivity are 1.0 mΩ cm, 2.1 mΩ cm, 2.0 mΩ cm, and 3.0 mΩ cm for the $YBa_2Cu_3O_{7-x}$ and $ErBa_2Cu_3O_{7-x}$ (Nos. 1 and 2—No. 2 reannealed), respectively.

Figures 1–3 also show the thermoelectric power of the three samples. The sign of S in the temperature interval of 77–300 K is positive for all three samples. Extrapolated values of S at 300 K, $S(300)$, are approximately 6.0 μV/K, 3.6 μV/K, and 3.0 μV/K for $YBa_2Cu_3O_{7-x}$, $ErBa_2Cu_3O_{7-x}$ (Nos. 1 and 2), respectively. The value of $S(300)$ of our $YBa_2Cu_3O_{7-x}$ sample is a little larger than that reported by Uher and Kaiser,[8] Kang *et al.*,[3] Laurent,[10] and Wang and Ong[11] but comparable to those reported by Trodahl and Mawdsley,[9] Mitra *et al.*,[16] and Cheong *et al.*[17] on polycrystalline samples, and those reported by Crommie *et al.*,[18] Forro *et al.*,[19] and Lu *et al.*,[20] on single crystals along the *ab* plane. A majority of results on polycrystalline[8,9,10,16] as well as single crystals[11,17–20] of $YBa_2Cu_3O_{7-x}$ indicate that $S(300)$ is positive. Exceptions are measurements reported by Lee *et al.*[5] and Khim *et al.*[21] for polycrystalline samples and Howson *et al.*[22] and Yu *et al.*[23] for measurements in the *ab* plane of single crystals. Our results confirm the positive sign of the thermoelectric power for $YBa_2Cu_3O_{7-x}$ and $ErBa_2Cu_3O_{7-x}$ samples prepared by a different method and thus confirm the earlier supposition that the majority carrier in $RBa_2Cu_3O_{7-x}$ superconductors are holes which therefore must be responsible for superconductivity in these materials. The only measurement of thermoelectric power on $ErBa_2Cu_3O_{7-x}$ oxide superconductors which is available in the literature is by Lee *et al.*[5] who report a negative room temperature thermopower of 3.4 μV/K. Although the magnitude of $S(300)$ of our $ErBa_2Cu_3O_{7-x}$ (No. 2) agrees, the sign does not agree with that reported by Lee *et al.*

Figure 1 shows the temperature dependence of S of $YBa_2Cu_3O_{7-x}$. It increases almost monotonically as T is lowered until T_c, where it drops sharply to zero. A small yet identifiable, peak is observed just above T_c. Very careful measurement in the transition region of the sample with ΔT values as small as 0.05 K did not show the presence of a strong peak as reported by Uher and Kaiser,[8] Trodahl and Mawdsley[9] (Gd-1:2:3 sample), and more recently Howson *et al.*[22] (single crystal). When the ends of the sample were not at precisely the same temper-

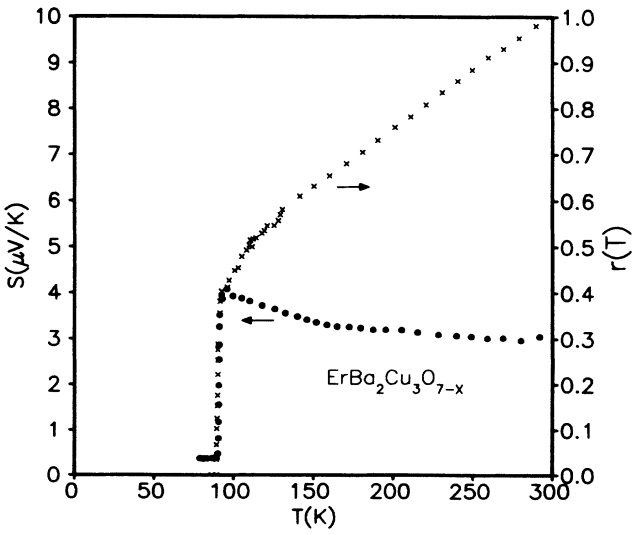

FIG. 3. Temperature dependence of the thermopower and the resistance ratio of $ErBa_2Cu_3O_{7-x}$ (No. 2).

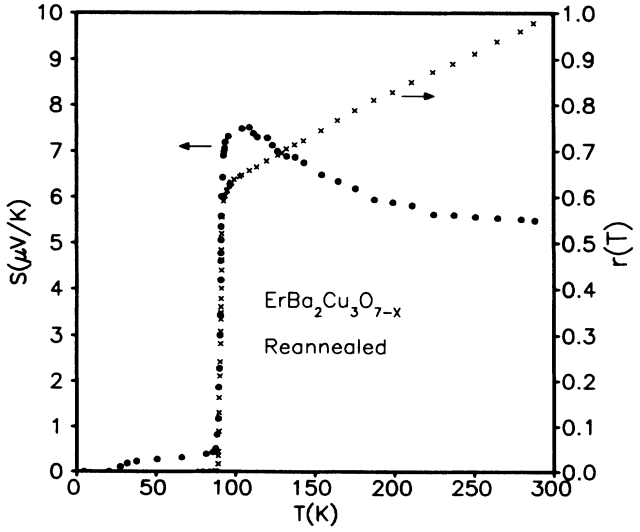

FIG. 4. Temperature dependence of the thermopower and the resistance ratio of $ErBa_2Cu_3O_{7-x}$ (No. 2) after reannealing in flowing oxygen at 500 °C for 12 h.

ature prior to the start of the heating period, we were able to generate a sharp, but totally artificial, peak in the transition region somewhat similar to the structure observed by Howson *et al.*[22] The behavior of S versus T for $YBa_2Cu_3O_{7-x}$ observed here is similar to that reported by Cheong *et al.*,[17] Mitra *et al.*,[16] Lu *et al.*[20] (single crystal), Trodahl and Mawdsley,[9] and Yu *et al.*[7] There is some structure present in S versus T data which we believe cannot be ignored and has been observed by others.[17]

Figures 2 and 3 show thermopower data for the two $ErBa_2Cu_3O_{7-x}$ samples. The sign of S is positive for both throughout the temperature interval. The temperature dependence of S is much weaker for these samples in comparison with that of $YBa_2Cu_3O_{7-x}$. Sample No. 2 shows a weaker temperature dependence than that shown for No. 1. Although the slope dS/dT is unquestionably small, it is clear that at higher temperatures it is negative and slowly increases in magnitude as T decreases. There is also evidence of a small peak just above T_c as was observed with $YBa_2Cu_3O_{7-x}$. The thermopower drops drastically at T_c (as defined by the resistance measurement) to a value of approximately 0.35 μV/K rather than zero. This has been observed in both $ErBa_2Cu_3O_{7-x}$ samples as well as for No. 2 after reannealing as discussed below. Data in the transition region was very reproducible and no hysteresis was found. The thermopower for No. 1 was zero below 30 K but then increased to approximately 0.35 μV/K above that temperature as if there were another superconducting transition. We shall comment on this behavior later.

Figure 4 shows data on $r(T)$ and $S(T)$ for $ErBa_2Cu_3O_{7-x}$ No. 2 after it was reannealed at 500 °C for 12 h in flowing oxygen. Reannealing did not change the sign of $dr(T)/dT$, nor did T_c change. However, the value of $r(0)$ changed significantly. A least-squares linear fit to the data gives $r(0)=0.472$ almost twice the value for the *as prepared* sample. The slope $dr(T)/dT$ also became smaller, 0.00176 K^{-1}. The value of $S(300)$ increased to 5.5 μV/K from 3 μV/K for the *as prepared* sample. The temperature dependence of S also shows a stronger variation below 200 K. The general trend of the S versus T data is similar to that observed by Wang and Ong[11] for the *ab* plane of single crystal $YBa_2Cu_3O_{7-x}$. At T_c,S drops sharply to approximately 0.35 μV/K and then goes to zero at about 25 K. Thus, the effect of reannealing sample No. 2, at least under the conditions mentioned, is to make it *less* metallic, i.e., to increase $r(0)$, to decrease dr/dT, and to increase the magnitudes of S and dS/dT.

DISCUSSION AND CONCLUSIONS

The thermopower and resistance ratio of the Y-Ba-Cu-O sample are comparable in magnitude and temperature dependence to values reported for a large number of polycrystalline samples[9,16,17] prepared by standard cold-pressed techniques. They are also similar to measurements in the *ab* plane made on several single-crystal samples.[18–20] (It should be noted that the temperature dependence of S along the c direction is much different from that in the ab-plane.)[11,20] This is not too surprising since thermal conductivity measurements[24] on Y-Ba-Cu-O samples prepared by this same hot-press sintering technique indicated an anisotropy with respect to the direction of pressure. Electron micrographs of the samples indicated that the hot-press sintering technique produced preferential alignment of the c axis of the grains along the direction of pressure. Our samples were cut with the long dimension perpendicular to the direction of pressure. Consequently, our samples would have some preferential orientation of the crystallites, with the ab plane along the direction of the induced thermoelectric field, but are otherwise equivalent to other polycrystalline R-Ba-Cu-O superconductors.

We also note that the behavior of $S(T)$ and $r(T)$ is very similar for the Y-based and the Er-based samples even though there is one distinct difference between these materials: Y is nonmagnetic; whereas, the rare-earth atoms, including Er, show local moments[25] above T_c. Malik *et al.*[26] have determined from susceptibility measurements that the effective moment per Er ion in $ErBa_2Cu_3O_{7-x}$ is $9.52\mu_B$, close to the theoretical value of $9.59\mu_B$ for the Er^{3+} ion. They also found evidence for antiferromagnetic interactions between them which has also been confirmed by neutron diffraction experiments.[27] Thus, the possibility for a contribution S_m to the thermopower for the Er-based samples due to magnon drag exists. S_m is generally much smaller than the phonon drag contribution to the thermopower even in magnetic materials. The similarity between $S(T)$ for the Y-based and the Er-based samples suggests that magnon drag effects are not important.

Another difference between the Y-based sample and the Er-based samples is that the thermopower for the Er-based samples (Nos. 1 and 2—2 reannealed) does not go to zero at the zero value of the resistive transition. We interpret this as evidence of diffusion or a chemical reaction with the Ag epoxy at the contact region into the $ErBa_2Cu_3O_{7-x}$ to form a doped region (presumably with Ag) with a significantly lower value of T_c (25–30 K). Since the leads to measure the thermal emf correspond to the current leads, this doped region would be in series with the remainder of the sample and produce a nonzero thermopower if it were not superconducting. For the resistance measurement this doped region is at the potential lead which draws no current; consequently, the fact that it is not superconducting will not be observed. Two possibilities can be offered to explain why no effect of the contacts on the thermopower for the Y-based sample was observed: (1) there is much less diffusion (reaction) from the epoxy into the $YBa_2Cu_3O_{7-x}$ at the curing temperature (75 °C) or, (2) the T_c for the doped region at the contact with $YBa_2Cu_3O_{7-x}$ is very close to that of the pure compound. It would be very interesting to determine if doping of $ErBa_2Cu_3O_{7-x}$ with Ag reduces T_c dramatically in comparison with similar doping of $YBa_2Cu_3O_{7-x}$. We also note that no nonzero values of thermopower below T_c have been observed for several Tl-based high-T_c superconductors that have been mounted and measured in an identical fashion in our laboratory.[28]

The literature values reported for the electrical transport coefficients, particularly the thermopower, of polycrystalline $R\mathrm{Ba_2Cu_3O_{7-x}}$ samples show a wide variation[3-11,16-23] which has often been attributed to variations in oxygen deficiency x, different anisotropy averages or varying influences from regions between crystalline grains. We note, however, that a similar wide variation is reported for measurements on single-crystal samples along a specified direction,[18-20,22,23] even differences in the reported sign of the thermopower. This suggests that the variation of transport properties is dominated by small variations in x which have no appreciable effect on T_c. Although the value of x was not determined independently, we observe a correlation between changes in $r(0)$, dr/dT, and S for our $\mathrm{ErBa_2Cu_3O_{7-x}}$ samples. Sample No. 2 has a smaller value of $r(0)$ and a larger value of dr/dT than sample No. 1 and, correlated with that observation, a smaller value of $S(300)$. When sample No. 2 was reannealed $r(0)$, $S(300)$, and the magnitude of dS/dT increased while dr/dT became smaller. Thus, there is a definite correlation between these four quantities.

This correlation between resistive and thermoelectric behavior in $R\mathrm{Ba_2Cu_3O_7}$ seems to be quite opposite to what one expects from a simple model of uncorrelated electrons (noninteracting, degenerate electron gas) as the majority carrier in a metallic system. The diffusion thermopower in this model is given by the Mott formula

$$S=\frac{\pi^2 k_B^2 T}{|e|}\frac{\partial \ln\sigma}{\partial\epsilon}\Big|\epsilon_F \ , \tag{1}$$

where any phonon or magnon drag terms have been ignored. The sign of S gives the sign of the majority carriers, and its magnitude is proportional to the density of states at the Fermi level. In this model S should decrease in magnitude with T for either type of carrier, hole, or electron. The same model also predicts the temperature dependence of S for a semiconductor or a material having a mobility gap at the Fermi level given by[29]

$$S=\frac{k_B}{|e|}\left[\frac{\Delta}{T}+B\right], \tag{2}$$

where Δ is the effective gap and B is a constant. In this case the magnitude of S will decrease with increasing T, but then the resistivity should increase with decreasing T. The temperature dependence of the resistivity and thermopower of our measurements as well as those reported for many other $R\mathrm{Ba_2Cu_3O_{7-x}}$ samples thus do not agree with the simple model of uncorrelated electrons. In addition, we find that S for the $\mathrm{ErBa_2Cu_3O_{7-x}}$ samples hardly shows any temperature variation between 200 and 300 K.

Anderson[30] has suggested that the Hubbard model provides an explanation for transport in $R\mathrm{Ba_2Cu_3O_{7-x}}$ alloys. In the high temperature, strong correlation limit (k_BT much greater than the band width W but much less than the on-site Coulomb repulsion U), the thermopower is dominated by the entropy of distribution of the charge carriers among available sites as given by the modified Heikes formula,[7,31]

$$S=-\frac{k_B}{|e|}\ln 2-\frac{k_B}{|e|}\ln\frac{1-n}{n} \ , \tag{3}$$

where n is the number of electrons per Cu site. This appears to be the only model capable of predicting a temperature independent thermopower. In $R\mathrm{Ba_2Cu_3O_{7-x}}$ there are two different Cu sites, corresponding to the planes and the chains. If we assume that these sites have the same valence, then $n=2(1+x)/3$. With this assumption and measured values of $S(300)$ the values of x for the Y-based sample and the Er-based samples, (Nos. 1, and 2— No. 2 reannealed) are 0.023, 0.015, 0.013, and 0.021, respectively. There are very small values, probably smaller than the actual values of x. Yu *et al.*[7] found that this model with strong Coulomb repulsion and charge carriers with spin seems to give the best fit to values of $S(300)$ for $\mathrm{YBa_2Cu_3O_{7-x}}$ samples as a function of x. This has led to the belief that correlated hopping of electrons from one Cu^{+2} to a nearby Cu^{+3} ion is responsible for the almost temperature independent thermopower for these compounds. Their recent measurements on single crystal $\mathrm{YBa_2Cu_3O_{7-x}}$ samples,[23] however, indicated that S in the Cu-O planes is small, negative, and almost temperature independent. Their results[23] also show that the thermopower of a $\mathrm{Sr_{0.15}La_{1.85}CuO_{4-x}}$ polycrystalline sample is independent of magnetic field up to 30 T. Thus, the spin-entropy contribution, the first term in Eq. (3), cannot be quenched even at such large values of H/T. They conclude from these results that the carriers in oxide superconductors do not have a free-spin degree of freedom; consequently, only models in which the carriers are bosons,[30] either lacking spin or moving as pairs with total spin zero, are appropriate.

Fisher *et al.*[32] have obtained a similar expression for the thermopower within a narrow band model. The high temperature limit is given by

$$S=\frac{k_B}{|e|}\ln\frac{1+x}{1-x} \ . \tag{4}$$

Values of x determined from values of $S(300)$ with this expression are of the same order as those determined from (3). Neither of these models predicts the observed temperature dependence of S (i.e., its increase below 100 K nor the correlations with $r(0)$ and dr/dT observed for most $R\mathrm{Ba_2Cu_3O_{7-x}}$ superconductors, and transport in these compounds remains unexplained. The possibility that the observed behavior in polycrystalline samples is a sum of contributions from semiconducting (in the regions between grains) and metallic behaviors in the crystallites appropriately weighted by the Gorter-Nordheim rule cannot be ruled out, but it would be difficult to explain the similar behavior observed for single crystals on that basis.

Mention should be made of recent calculations of transport coefficients for single crystal $\mathrm{YBa_2Cu_3O_{7-x}}$ materials by Allen *et al.*[33] based on band theory. They predict that S_{xx} and S_{yy} (in the ab plane) should be negative and that the sign of S_{zz} depends on the choice of the energy dependence of the scattering time τ. Our measurements on these preferentially oriented hot-pressed samples disagree in sign with the predictions by Allen *et al.*,[33]

as do measurements in the *ab* plane for single-crystal samples by Crommie *et al.*,[18] Forro *et al.*,[19] and Lu *et al.*,[20] but the temperature dependence is similar to that of the predictions. The most striking prediction of this theory is that the Hall coefficient changes sign as the field is rotated from along the *c* axis to lie in the ab plane, in good agreement with measurements by Tozer *et al.*[34] The predicted room temperature values of ρ_{xx} and ρ_{xy}, 37 and 16 $\mu\Omega$ cm, are much smaller than any values observed for either single crystal or polycrystalline $YBa_2Cu_3O_{7-x}$.

Anderson and Zou[35] observed that a plot of $T\rho_{ab}$ was linear against T^2 for single-crystal $YBa_2Cu_3O_{7-x}$ samples.[34] Within the framework of resonating-valence-bond theory[36] they have suggested the following relationships between ρ and T for single crystal $R Ba_2Cu_3O_{7-x}$ samples

$$\rho_{ab} = A_{ab}/T + B_{ab}T \tag{5}$$

and

$$\rho_c = A_c/T + B_cT \ . \tag{6}$$

Hagen *et al.*[37] found good agreement between their experimental data and these equations. We have fitted our high temperature ($T > 200$ K) data on these polycrystalline samples to a relation of the type above and also to the usual linear relationship for metallic conductivity. The standard deviation for the Anderson-Zou relation was a factor of three larger, but this does not constitute a rigorous test of the theory.

We conclude that a good theoretical understanding of electrical transport in the normal state of these superconducting oxides is lacking.

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