Polaron conductivity mechanism in $PrBa_2Cu_3O_{6.9}$

S. V. Varyukhin and O. E. Parfenov

Kurchatov Institute Russian Science Center, 123182 Moscow, Russia

(Submitted 4 November 1993)
Pis'ma Zh. Eksp. Teor. Fiz. **58**, No. 11, 889–892 (10 December 1993)

The temperature dependence of the ac conductivity $\sigma=\sigma_0+\sigma(\omega)$ of a $PrBa_2Cu_3O_{6.9}$ sample at low frequencies has been studied. The term σ (ω) has an anomalous temperature dependence at low temperatures. The feature observed is due to a polaron conductivity mechanism.

The discovery of superconductivity in metal oxides raised several questions concerning the mechanisms for the formation of a metallic state in compounds whose primary distinction from conventional metals and semiconductors was a predominantly ionic bonding. The pronounced localization of charges, including impurity carriers, weakens the screening of the Coulomb interaction in such systems in comparison with systems with covalent bonds. As a result, the local Coulomb perturbation causes a substantial deformation of the nearest neighborhood and gives rise to polaron states.[1] This circumstance has served as the basis for models which treat the formation of (bi-) polaron states as one possible mechanism for the high-T_c superconductivity of the metal oxides.[2-7] A study of polaron characteristics in the metallic state is hindered by the renormalization of the original states due to the high carrier concentration. In the insulating phase, however, the formation of polaron states strongly influences the nature of the conductivity at low temperatures. In a region in which the transport occurs through hops of carriers between localized states, the ac conductivity is

$$\sigma=\sigma_0+\sigma(\omega)=\sigma_0+A\omega^s, \tag{1}$$

where $\omega=2\pi f$ is the measurement frequency, and the exponent s is $\leqslant 1$ (Ref. 1). Various conductivity mechanisms lead to different forms of the dependence of the term $\sigma(\omega)=A\omega^s$ (Refs. 8 and 9). In the case in which polaron states form, the exponent s is observed to have an anomalous temperature dependence.[8] Our purposes in the present study were to measure the temperature dependence of the ac conductivity at various measurement frequencies, to single out the term $\sigma(\omega)=A\omega^s$, and to analyze the results of the $s(T)$ measurements on the basis of existing theories.

In the present experiments we studied the compound $PrBa_2Cu_3O_y$, which is one of the few exceptional cases among the 1-2-3 systems based on other lanthanides in that it has yet to be produced in a either a superconducting or metallic state. The reason, as was shown in Ref. 10, is a pronounced localization of carriers. This circumstance distinguishes $PrBa_2Cu_3O_y$ as a compound in which polaron effects should be seen most clearly. For the experiments we prepared a polycrystalline sample with an oxygen concentration close to the maximum value attainable, $y=6.9$. The conductivity measurements were carried out over the frequency range $0.3 \leqslant f \leqslant 100$ kHz in

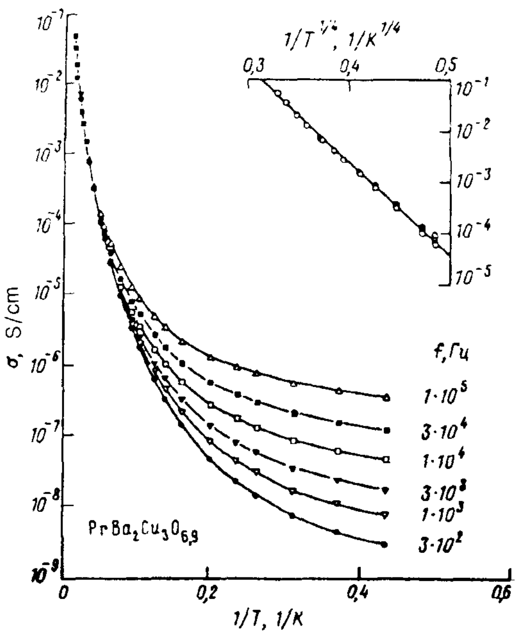

FIG. 1. Temperature dependence of the conductivity σ at six measurement frequencies between 300 Hz and 100 kHz. The inset shows a part of a σ (T) curve on which there is no dependence on the measurement frequency, in the coordinates $\log\sigma$, $1/T^{1/4}$.

the temperature range $1.1 < T < 85$ K. The procedure for preparing the compound, the procedure for preparing the test sample for the measurements, and the experimental procedures are described in Refs. 10 and 11.

Figure 1 shows results of measurements of the temperature dependence of the conductivity σ for six measurement frequencies. We see that there is essentially no frequency dependence at temperatures $T > 20$ K, and the conductivity is described approximately by $\sigma \propto \exp -(\Delta/T)^{1/4}$ (see the inset in Fig. 1). This result is evidence of a hopping conductivity, which leads to a significant frequency dependence at lower temperatures (Fig. 1). Figure 2a shows the frequency dependence of σ at various temperatures. The curves are drawn in accordance with (1), which holds over the entire frequency range studied. Figure 2b shows the frequency dependence with the term $\sigma(\omega) = \sigma - \sigma_0 = A\omega^s$. We see that the exponent s (the slope of the straight lines) is a nonmonotonic function of the temperature. A monotonic decrease in the exponent s with increasing temperature has been observed[12] previously for the case of $LaCuO_{4+y}$. Figure 3 shows the entire $s(T)$ dependence for $PrBa_2Cu_3O_{6.9}$. The structural feature observed on the $s(T)$ curve—a minimum as a function of the temperature—can be described in a model with a tunneling of large-radius polarons, for which an overlap of regions of lattice distortion is important.[8] The height of the effective barrier, W, depends on the distance (R) over which an electron must be transported:

$$W = W_p(1 - r_p/R), \tag{2}$$

where the polaron energy is

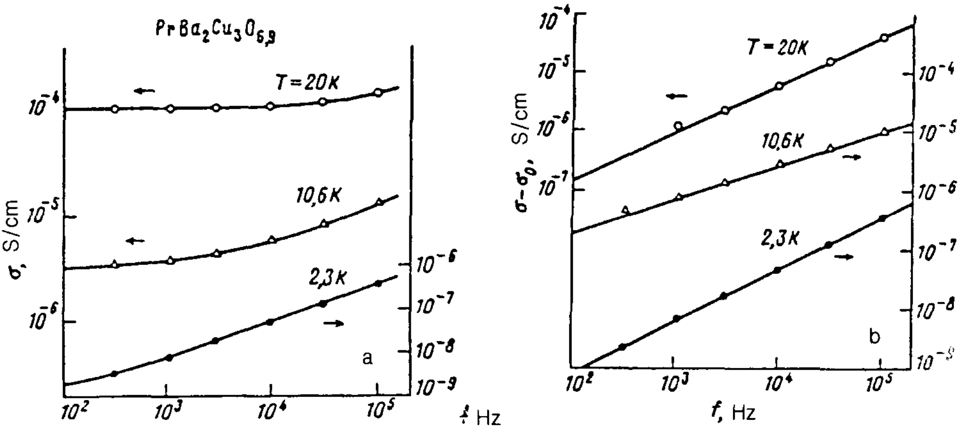

FIG. 2. Frequency dependence of the conductivity σ (a) and $\sigma(\omega)=\sigma-\sigma_0$ (b) for three temperatures. The curves in frame a are drawn in accordance with Eq. (1).

$$W_p=e^2/4\epsilon_p r_p, \tag{3}$$

r_p is the radius of the polaron, $\epsilon_p^{-1}=\epsilon_\infty^{-1}-\epsilon_0^{-1}$ is the effective dielectric constant, and ϵ_∞ and ϵ_0 are the optical and static dielectric constants. The temperature dependence of s is described by[8]

$$P^2+[W_p/kT+\ln(\omega\tau_0)]P-W_p r_p'/kT=0, \tag{4}$$

$$(1-s)P=\frac{4+6(W_p r_p'/P^2kT)}{[1+(W_p r_p'/P^2kT)]^2}, \tag{5}$$

where $P=2\alpha R_\omega$, $r_p'=2\alpha r_p$, R_ω is a characteristic hopping length for the given temperature, the parameter α characterizes the size of the electron wave function of the localized state, and τ_0^{-1} is a characteristic frequency of optical phonons. Equations (4)

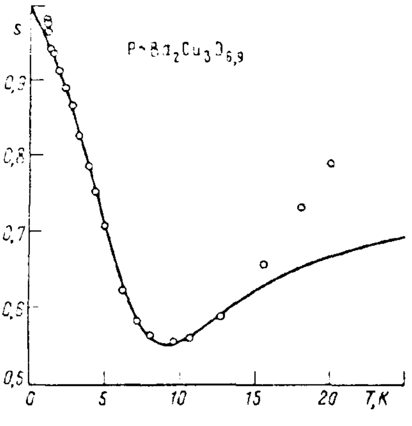

FIG. 3. Temperature dependence of the factor s. The curve is drawn in accordance with Eqs. (4) and (5) with the parameter values $\ln(\omega\tau_0)=-18$, $r_0'=1.75$, and $W_p=150$ K.

and (5) describe a series of $s(T)$ curves with a minimum. The basic parameters of these curves are r'_p and W_p, which determine the value of $s(T)$ at the minimum and its position along the temperature scale, respectively. Figure 3 shows a curve drawn in accordance with Eqs. (4) and (5) with the parameter value $\ln(\omega\tau_0) = -18$ (which corresponds to $\tau_0 \simeq 10^{-13}$ s for the measurement frequencies used), $r'_p = 1.75$, and $W_p = 150$ K. The deviation of the experimental points from the curve at $T > 15$ K apparently means that at high temperatures it is not strictly valid to use the dipole approximation to describe the transition of particles between two isolated centers, within the framework of which expressions (4) and (5) were derived. Transitions among three and more sites become important. An increase in the probability for the motion of particles along chains of sites leads to an increase in the term σ_0 in (1), which describes the dc conductivity. This result can be seen from Fig. 2a, where we have $\sigma_0 \ll \sigma(\omega)$ at $T = 2.3$ K, while at $T = 20$ K we find the opposite situation: $\sigma_0 \gg \sigma(\omega)$, with a total conductivity $\sigma \simeq \sigma_0$. Accordingly, the parameters of the approximation were chosen for best agreement in the low-temperature region (Fig. 3). Using the value found for W_p from expression (3), we can estimate the polaron radius r_p. For $PrBa_2Cu_3O_y$ with $y = 6.9$, using $\epsilon_\infty \simeq 2$ (Ref. 13) and $\epsilon_0 \simeq 25$ (Ref. 10), we find $r_p = 125$ Å.

This large value of r_p apparently cannot be accepted as a characteristic of an individual polaron formed when charge is introduced into the $PrBa_2Cu_3O_y$ matrix. In the first place, the model[8] proposed to explain the features of $s(T)$ corresponds to the isotropic case, while the 1-2-3 systems have a clearly expressed 2D conductivity, resulting from the presence of conducting CuO_2 planes. Second, the high carrier concentration[11] in $PrBa_2Cu_3O_{6.9}$ should lead to a pronounced overlap of regions of lattice distortions and thus a renormalization of Eq. (2), derived for two remote states between which an electron is transported.

We wish to thank G. N. Chubchev for assistance in analyzing the experimental results. This study was carried out as part of project 90347 of the State Program on High Temperature Superconductivity.

[1] N. F. Mott and E. A. Davis, *Electronic Processes in Non-Crystalline Materials* (Oxford Univ. Press, New York, 1979).
[2] D. Emin, Phys. Rev. B **45**, 5525 (1992).
[3] N. F. Mott and A. S. Alexandrov, Supercond. Sci. Technol. **6**, 215 (1993).
[4] J. Ranninger, Z. Phys. B **84**, 167 (1991).
[5] R. C. Baetzold, Phys. Rev. B **42**, 56 (1990).
[6] S. Robaszkiewicz, R. Micnas, and J. Ranninger, Phys. Rev. B **36**, 180 (1987).
[7] G. Verbist, F. M. Peeters, and J. T. Deureese, Phys. Rev. B **43**, 2712 (1991).
[8] A. R. Long, Adv. Phys. **31**, 553 (1982).
[9] S. R. Elliott, Adv. Phys. **36**, 135 (1987).
[10] S. V. Varyukhin and O. E. Parfenov, JETP Lett. **58**, 101 (1993).
[11] O. E. Parfionov and A. A. Konovalov, Physica C **202**, 385 (1992).
[12] C. Y. Chen, R. J. Birgeneau, M. A. Kastner *et al.*, Phys. Rev. B **43**, 392 (1991).
[13] J. Kircher, M. Cardona, S. Goplan *et al.*, Phys. Rev. B **44**, 2410 (1991).

Translated by D. Parsons