Low-frequency conductivity and polaron transport in the dielectric phase of YBaCuO

S. V. Varyukhin and O. E. Parfenov

Kurchatov Institute, 123182 Moscow, Russia
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The temperature dependence of the ac conductivity $\sigma=\sigma_0+A\omega^s$ of the sample $YBa_2Cu_3O_{6.2}$ was investigated at low frequencies. An anomalous temperature dependence of the term $A\omega^s$ was found: specifically, $s(T)$ is observed to have a minimum. It was shown that the observed singularity can be explained on the basis of a model that assumes tunneling of large-radius polarons, for which the overlap of regions of lattice distortion is large. Estimates are obtained for the radius and binding energy of a polaron. © *1995 American Institute of Physics.*

The formation of (bi)polaron states in metal oxides is considered to be a possible mechanism for high-T_c superconductivity.[1,2] The validity of this approach to the problem of high-T_c superconductivity is based on the fact that metal oxides are systems in which ionic coupling predominates, in Coulomb screening is substantially attenuated and the local Coulomb perturbation gives rise to a significant deformation of the lattice and the formation of polaron states.[3] The experimental discovery of features in the IR conductivity spectra can be explained on the basis of models which presuppose polaron conductivity.[4] Analogous singularities were observed in CuO, which is a fundamental unit of high-temperature superconductors.[5] On the other hand, the existence of low-mobility carriers, such as polarons, leads to the fact that in the temperature range of the hopping mechanism of conduction, the probability of tunneling between isolated localized states is much lower than in the case of electrons. As a result, at low frequencies the conductivity is observed to depend on the frequency. In the general case, the frequency dependence has the form

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

where $s<1$.[3] The term $\sigma(\omega)=A\omega^s$ is determined by both the type of carrier and the transition mechanism of the between localized states; in the case of polaron transport the factor $s(T)$ has a pronounced anomaly — a temperature minimum, whose position is determined by the binding energy of the polaron states. The theories describing ac conductivity in the hopping mechanism of conduction are reviewed in detail in Refs. 6 and 7.

In Ref. 8 we observed a feature in $s(T)$ for $PrBa_2Cu_3O_{6.9}$. This agrees with the predictions of the theory of Ref. 6, but an attempt to estimate the polaron radius gave an unphysical result — the polaron radius was much greater than the average intercarrier distance. This is a consequence of the fact that in the compound $PrBa_2Cu_3O_y$ with $y=6.9$, which was studied in Ref. 8 and has a much larger number of carriers than the dielectric composition with $y=6.0$, the regions of lattice distortions which form the polaron cloud overlap, and this should result in a substantial renormalization of the relations obtained for isolated, widely separated polaron states. As noted in Ref. 8, if the relations obtained for separate polaron states are used to estimate the polaron radius in systems with a high carrier concentration, the result may be too high, and to study the characteristics of polaron states, it is necessary to investigate compounds with a low carrier concentration.

In the present paper we present measurements of the low-frequency conductivity of a dielectric sample of $YBa_2Cu_3O_y$ with $y=6.2$. The experimental composition was prepared using the standard ceramic technology with sintering followed by annealing in order to obtain the required oxygen concentration. The oxygen concentration in the samples was changed by using different regimes of annealing followed by quenching in a helium atmosphere.[9] The nonuniformity of the oxygen concentration over the volume of the sample was $\Delta y\leqslant 0.03$; it was investigated by means of x-ray diffraction and estimated according to the width of the Bragg peaks. The sample consisted of a pellet $\simeq 4.5$ mm in diameter and $\simeq 0.8$ mm thick, on both sides of which silver was sputtered. Silver was used because in order to measure small conductivities, an ohmic contact must be obtained between the metal and the dielectric sample. Alloying makes it possible to obtain a low-resistance ohmic contact between the silver and the experimental dielectric compound. Heat treatment can be applied to the sample after deposition of the contact; there are definite advantages to this procedure, because after annealing the oxygen content in the contact region does not change and corresponds to the volume concentration. The low-frequency impedance of the flat capacitor thus obtained was investigated in the frequency range $0.3\leqslant f\leqslant 100$ kHz ($\omega=2\pi f$) in the temperature range $1.5<T<300$ K. The flat-capacitor geometry is used because it is necessary to eliminate any effect of the parasitic capacitance of the sample surface, which can substantially distort the results for small values of the conductivity of dielectrics at low temperatures. The amplitude and phase of the current flowing through the sample were measured by measuring the voltage across 500 Ω resistor in series with the experimental sample. The effect of the capacitance of the feed cables was taken into account in the calculation of the conductivity.

Figure 1 displays the temperature dependence $\log(\sigma)$ versus $1/T$ for six frequencies at which measurements were performed. One can see that in the region $T<150$ K, σ depends strongly on the frequency ω. The frequency dependence of σ at different temperatures is shown in Fig. 2. The curves were plotted according to the relation (1), which holds over the entire experimental frequency range. Figure 3

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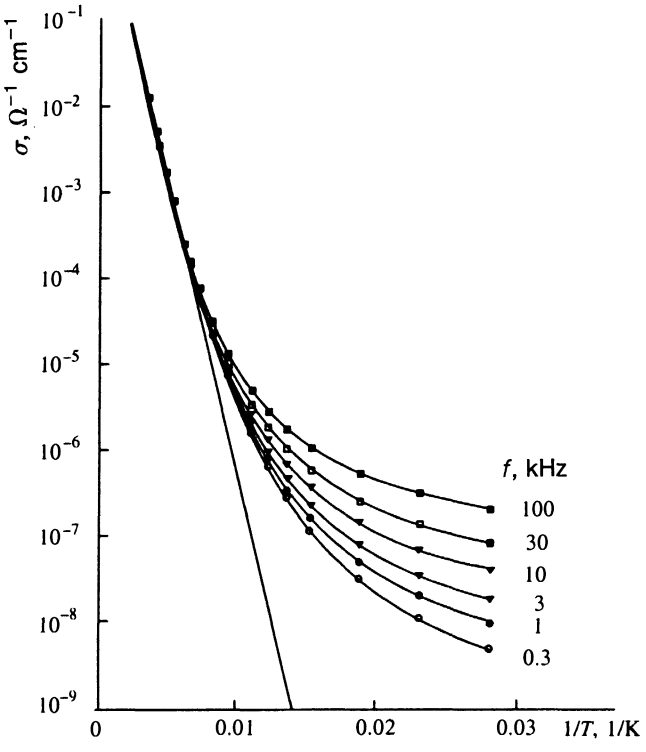

FIG. 1. Temperature dependence of the conductivity σ of $YBa_2Cu_3O_{6.2}$ for six different frequencies ranging from 0.3 Hz to 100 kHz.

displays the frequencies of the term $\sigma(\omega)=\sigma-\sigma_0=A\omega^s$. One can see that the exponent s (the slope of the straight lines) is a nonmonotonic function of the temperature. The entire temperature dependence $s(T)$ is displayed in Fig. 4. The observed of the function $s(T)$, a temperature minimum,

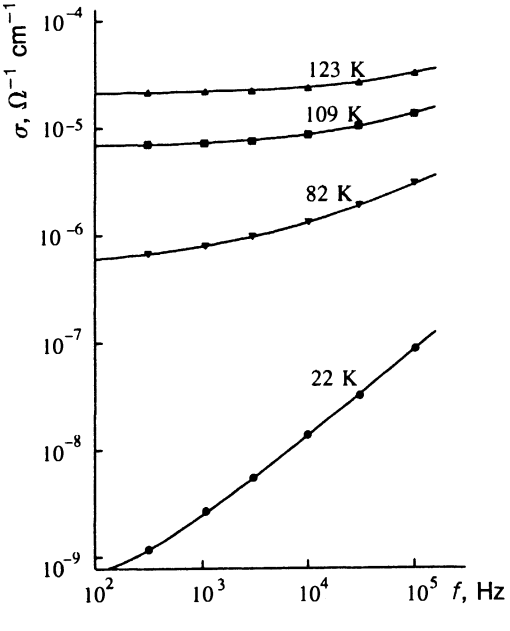

FIG. 2. Frequency dependence of the conductivity σ of the compound $YBa_2Cu_3O_{6.2}$ at different temperatures. The curves are plotted according to the relation (1).

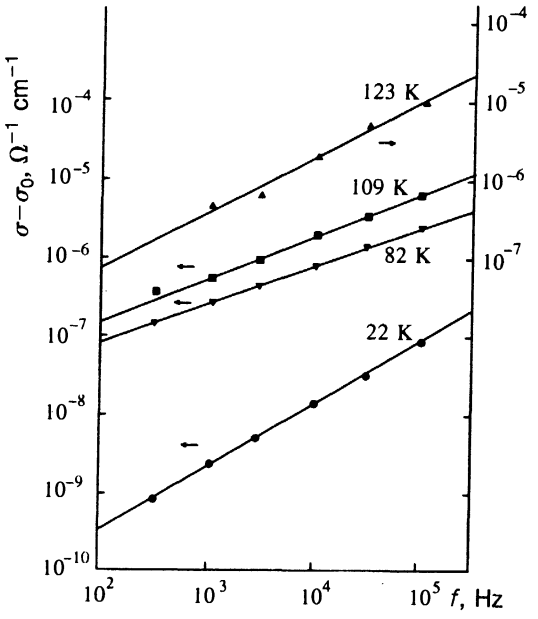

FIG. 3. Frequency dependence of the term $\sigma(\omega)=\sigma-\sigma_0=A\omega^s$ in the expression (1).

can be described on the basis of a model that assumes tunneling of large-radius polarons, for which there is a significant overlap of the regions of lattice distortion.[6] The height W of the effective barrier depends on the distance R to which the electron must be transported

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

where the polaron energy is

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

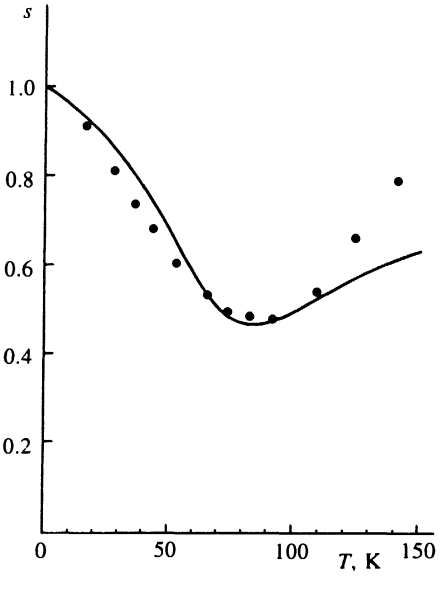

FIG. 4. Temperature dependence of the exponent s in the expression (1) for $YBa_2Cu_3O_{6.3}$. The curve is plotted according to (4)–(5) of Ref. 6.

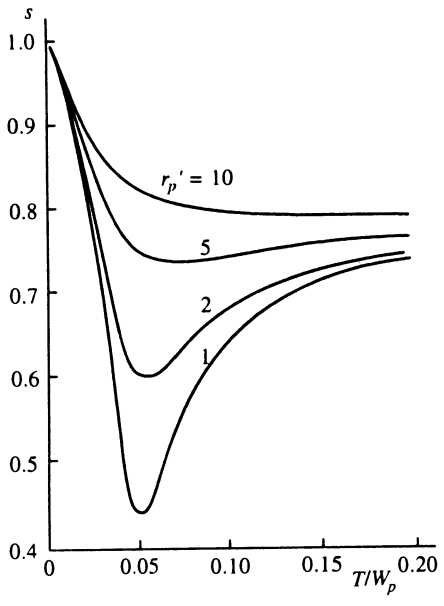

FIG. 5. Temperature dependence of the factor s for different values of the polaron radius.[6]

r_p is the radius of the polaron, the effective dielectric constant $\epsilon_p^{-1}=\epsilon_\infty^{-1}-\epsilon_0^{-1}$, and ϵ_∞ and ϵ_0 are the optical and static dielectric constants, respectively. The temperature dependence of the factor s is described by the relations[6]

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

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

where $P=2\alpha R_\omega$, $r_p'=2\alpha r_p$, R_ω is the characteristic hopping distance at a given temperature, α is a parameter characterizing the extent of the electron wave function of a localized state, and τ_0^{-1} is the characteristic frequency of the optical phonons. Equations (4) and (5) describe a series of functions $s(T)$ with a minimum. These functions are displayed in Fig. 5. The main parameters are r_p' and W_p, which determine the values of $s(T)$ at the minimum and the temperature at the minimum, respectively. Figure 4 displays the curve plotted according to (4) and (5) with $\ln(\omega\tau_0)=-18$ (which corresponds to $\tau_0\simeq 10^{-13}$ sec for the measurement frequencies employed), $r_p'=1.1$, and $W_p=1700$ K. The deviation of the experimental points from the curve observed at $T>110$ K probably due to the fact that at sufficiently high temperatures, the dipole approximation [within which (4) and (5) were obtained], describing the passage of a particle between two isolated centers, does not hold exactly. Transitions between three or more sites start to have a large effect. As the probability of motion of particles along chains of sites increases, the term σ_0, describing the dc conductivity, in Eq. (1) increases. This can be seen in Fig. 2, where $\sigma_0 \ll \sigma(\omega)$ at $T=22$ K; at $T=123$ K the opposite is observed: $\sigma_0 \gg \sigma(\omega)$ and the total conductivity $\sigma \cong \sigma_0$.

Using (3) and $\epsilon_\infty \simeq 3$ (Ref. 10) and $\epsilon_0 \simeq 15$ (Ref. 11) for $YBa_2Cu_3O_y$, the radius of the polaron states can be estimated to be $r_p \simeq 6.5$ Å. This estimate of r_p agrees with the model employed to explain the observed temperature dependence $s(T)$, since the region of polaron distortion of the lattice $\sim 2r_p$ is much greater than the cell parameters a and b of the 1-2-3 structure in the CuO plane whichs confirms the validity of the model employed. The latter assumes the existence of large-radius polarons. It should be noted, however, that $c \sim 2r_p$, probably because the model of Ref. 6, proposed to explain the singularity of $s(T)$, corresponds to the isotropic case, while the conductivity of the 1-2-3 system has an obvious two-dimensional character, and the value obtained for r_p can be regarded only as a first approximation.

At high temperatures, where σ does not depend on ω, the temperature dependence $\sigma(T) \propto \exp(-\Delta/T)$ with $\Delta \simeq 1550$ K (Fig. 1, straight line). The closeness of the magnitudes of Δ and W_p suggests that the parameters of the polaron states do not change as the temperature T increases, and over the entire temperature range, the conductivity is determined by the presence of polarons with a binding energy of $\simeq 0.14$ eV. At low temperatures, the conductivity is determined by tunneling between localized states; this leads to a strong frequency dependence of the conductivity. At high temperatures, the main contribution to the conductivity comes from carrier delocalization, which is determined by the decay probability of a polaron state, and the temperature dependence of the conductivity then has a thermal activation character, $\sigma(T) \propto \exp(-W_p/T)$.

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[1] D. Emin, Phys. Rev. B **45**, 5525 (1992).
[2] N. F. Mott and A. S. Alexandrov, Supercond. Sci. Technol. **6**, 215 (1993).
[3] N. F. Mott and E. A. Davis, *Electronic Processes in Non-Crystalline Materials*, Clarendon Press, Oxford (1971).
[4] D. Mihailovic, C. M. Foster, K. Voss, and A. J. Heeger, Phys. Rev. B **42**, 7989 (1990).
[5] A. A. Samokhvalov, N. A. Viglin, B. A. Gizhevskiĭ, N. N. Loshkareva *et al.*, Zh. Éksp. Teor. Fiz. **103**, 951 (1993); JETP **76**, 463 (1993)].
[6] A. R. Long, Adv. Phys. **31**, 553 (1982).
[7] S. R. Elliott, Adv. Phys. **36**, 135 (1987).
[8] S. V. Varyukhin and O. E. Parfenov, Pis'ma Zh. Éksp. Teor. Fiz. **58**, 889 (1993) [JETP Lett. **58**, 830 (1993)].
[9] O. E. Parfionov and A. A. Konovalov, Physica C **202**, 385 (1992).
[10] J. Kircher, M. K. Kelly, S. Rashkeev, M. Alouani *et al.*, Phys. Rev. B **44**, 217 (1991).
[11] S. V. Varyuknin and O. E. Parfenov, Pis'ma Zh. Éksp. Teor. Fiz. **58**, 98 (1993) [JETP Lett. **58**, 101 (1993)].

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