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Kyoto University
The ac Conductivity of LaCuO₄

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The ac conductivity of LaCuO₄ and La₂₋ₓSrₓCuO₄ was measured. Conductivity and dielectric constant have little ω dependence in the temperature range above 40 K. A dielectric relaxation is found below 20 K. This is caused by the bound polaron. Dielectric constant decreases with applied voltage and conductivity increases with applied voltage. Tunneling motion of holes and bound polaron hopping would induce these results.

KEY WORDS: LaCuO₄ / La₂₋ₓSrₓCuO₄ / Dielectric constant / ac conductivity

Introduction

LaCuO₄ is an antiferromagnetic insulator. By introducing Sr ion in this compound the antiferromagnetism is destroyed and spin glass property appears. The superconductivity occurs in close vicinity to an insulator metal transition. Conductivity and dielectric constant have been measured for LaCuO₄₁₋ₓ. It is said that σ' and σ'' (ε) depend on ω: σ' = σ₀ + ωε₀ε₉tan(ω/2)σ₀ω. Hopping conductivity character has been observed in LaCuO₄. At higher frequency above 10⁶ Hz interfacial polarization caused by the contact between sample and electrode was found. In the lower frequency measurement (f<10⁵) the effect of the contact is not so large. In this work ac conductivity was measured in the frequency range from 10² th 10¹ Hz for sintered LaCuO₄ and La₂₋ₓSrₓCuO₄₋ₓ (x≤0.05) samples. The measurement was carried out in the temperature range from 4.2 to 100K. ω dependence of σ' and σ'' is examined and voltage dependence of σ' and σ'' is also studied. Relaxation due to the bound polaron hopping is studied.

Experimental

Disk samples of sintered LaCuO₄ and La₂₋ₓSrₓCuO₄₋ₓ were prepared by the following method. The mixture of La₂O₃ and SrCO₃ powders was calcined at 900°C in air. After pulverization and homogenization of the calcined samples the samples were pressed into disk form of 17 mm diameter and 2 mm thickness and sintered in the temperature range between 900-980°C in air. These processes were repeated 3 times. The surface of the sample was coated with Al electrode evaporated in vacuum or Ag electrode painted with Dotite paste. The disk sample was mounted on Al dielectric cell in a cryostat. Impedance measurement was carried out by HP 4274 A LCR meter.

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Results and Discussion

Several disk samples of LaCuO$_4$ were prepared by sintering at the temperature between 900-980°C in air. Dielectric constant and conductivity of the most representative sample is shown in Fig.1. Dielectric constant of $-10^4$ at 100 Hz is very high compared with that of our previous report$^1$ and other references$^{2,3}$ ($\varepsilon = 50$). Although conductivity of this sample is a little larger than that of the previous sample$^7$, it is smaller than that of other report$^{1,5}$. It seems that this sample has different hole concentration from that of other sample. At 4.2K dielectric constant will become $-500$ at 100 kHz because the dielectric relaxation is finished. Log $\varepsilon'$ - log f dependence and log $\kappa'$ - log f dependence show slightly bent curve because of a dielectric relaxation. At higher temperature (40.4K) $\omega$ dependence of dielectric constant and conductivity becomes smaller as shown in Fig.2. By the hopping model for LaCuO$_4$ the relation $\sigma^+ = \sigma_{dc} + \sigma_0 \omega^+$, $\sigma^- = \omega \varepsilon_0 \varepsilon \infty + \tan (s \pi/2) \sigma_0 \omega^+$ has been reported. But no clear $\omega$ dependence of dielectric constant and conductivity is not observed. LaCuO$_4$ shows the temperature dependence of dielectric constant and conductivity as shown in Fig.3 (a) and (b), respectively. Dielectric constant increases rapidly from 500 to 16000 at low temperature. Dielectric constant at 100 Hz becomes dispersed above 20 K because the sample has higher conductivity at these temperatures and the measurement becomes incorrect. Dielectric relaxation is found near 10 K. Activation energy of the relaxation is 0.0043 eV. This motion of charge carrier with very low activation energy may correspond to the motion of bound polaron$^1$. In reference 1 this effect remains even at room temperature near the frequency of $10^6$ Hz but the relaxation of this work becomes very fast and can not be observed above 30
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K. Impedance was measured under the voltages 0.029, 0.24 and 1.4 V/cm. At higher applied voltage of 1.44 V/cm the peak of conductivity becomes unclear (Figs. 4 (a) and (b)). This suggests that higher applied field changes the motion of the charge carriers in the lattice. Temperatue dependence of dielectric constant and conductivity of La$_{1.95}$Sr$_{0.05}$CuO$_{4-\delta}$ is shown in Fig. 5 (a) and (b), respectively. Dielectric constant decreases near 4.2 K, whereas it has almost same value in the temperature range from 20 K to 100 K. Conductivity at 100 kHz increases near 4.2 K corresponding to the decrease of dielectric constant in Fig. 5 (a). It seems that there is a dielectric relaxation near 4.2 K. It appears at lower temperature than that of LaCuO$_4$ sample. Generally it is expected that conductivity and dielectric constant of this sample should have higher values than those of LaCuO$_4$, because the concentration of hole which is the charge carrier in this compound is higher for La$_{1.95}$Sr$_{0.05}$CuO$_{4-\delta}$ . However, dielectric constant of this sample is $0.5 \times 10^4$ and conductivity $1 \times 10 \mu$S/cm, whereas those of LaCuO$_4$ in Fig. 3 are $1.6 \times 10^4$ and $10^2 - 10^3 \mu$S/cm. Dielectric constant and conductivity of LaCuO$_4$ are higher than those of La$_{1.95}$Sr$_{0.05}$CuO$_{4-\delta}$ . Therefore the sample of La$_{1.95}$Sr$_{0.05}$CuO$_{4-\delta}$ has not so many holes as LaCuO$_4$ in Fig. 3. It is suggested by the previous work that this type of dielectric relaxation appears at lower temperature for the samples with higher Sr concentration$^7$. This dielectric relaxation is considered to be caused by the bound polaron and its motion becomes faster for the samples with higher concentration of hole. In this compound (LaCuO$_4$) it is generally said that $\sigma'$, $\sigma''$ depend on $\omega$ and $\sigma$ shows only weak temperature dependence and hence tunneling motion of charge carriers between localized state is the main mechanism of transport$^1$. Variable range hopping is examined by the relation $\rho = \rho_0 \exp \left( \frac{(T_0/T)}{\gamma} \right)$ ($\gamma = 1/4$ for three dimensional and $\gamma = 1/3$ for two dimensional). But in the low temperature region of the sample of this work the examination of this relation can not be obtained because of the existence of the dielectric relaxation. In the temperature range above 40 K, $\gamma = 1/3$ fitting is good for the data in Fig. 3.
Fig. 3 (a) Temperature dependence of dielectric constant of La$_2$CuO$_4$. 
(b) Temperature dependence of conductivity of La$_2$CuO$_4$. The measured voltage is 0.029 V/cm. A: 0.1, B: 1, C: 10, and D: 100 kHz.
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Fig. 4 (a) Temperature dependence of dielectric constant of La$_2$CuO$_4$.
(b) Temperature dependence of conductivity of La$_2$CuO$_4$. The measured voltage is 1.4 V/cm. A: 0.1, B: 1, C: 10, and D: 100 kHz.
Fig. 5 (a) Temperature dependence of dielectric constant of La$_{1.95}$Sr$_{0.05}$CuO$_4$ - $\varepsilon$
(b) Temperature dependence of conductivity of La$_{1.95}$Sr$_{0.05}$CuO$_4$ - $\sigma$
The measured voltage is 0.081 V/cm. A: 0.1, B: 1, C: 10, and D: 100 kHz.
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Fig. 6 Temperature dependence of conductivity of $\text{La}_{1.96}\text{Sr}_{0.04}\text{CuO}_4$. The measured voltage is 0.0256 V/cm and frequencies are 0.1, 1, 10, and 100 kHz.

Fig. 7 Frequency dependence of dielectric constant, loss and conductivity of $\text{La}_2\text{CuO}_4$ at 4.2K. The measured voltage is 0.037 (A), 2.1 (B) and 3.88 (C) V/cm.
Figure 6 shows the temperature dependence of conductivity of La$_{1.96}$Sr$_{0.04}$CuO$_4$. This sample has large conductivity but dielectric relaxation near 4.2 K is not seen in this figure. It is considered that the sample with high conductivity has no dielectric relaxation or has the relaxation with very short time constant and can not be found in the temperature range from 4.2 to 100 K. Figure 7 shows the frequency dependence of dielectric constant, dielectric loss and conductivity of LaCuO$_4$ under the applied field (0.037, 2.1 and 3.88 V/cm). Although dielectric loss has a maximum near $10^3$ Hz ($\varepsilon'' = \frac{\varepsilon' (\omega) - \varepsilon' (100)}{\omega}$), real part $\varepsilon'$ does not shows the flat part in the low frequency limit of the Debye type relaxation. From Fig.3 relaxation frequency will be supposed to be below 1 kHz at 4.2 K. In Fig.7 most interesting result is that dielectric constant and conductivity are dependent on applied voltage. Dielectric constant decreases with applied voltage and conductivity increases with applied voltage. This tendency is prominent at low frequency but the difference of dielectric constant and conductivity between applied fields becomes little at high frequency. The dc conductivity increases with applied field, whereas dielectric displacement of charge carrier decreases with applied field. Non ohmic resistance of these kinds of material suggests the character of charge carrier holes. The number of holes with the tunneling motion becomes large with the field but that with high frequency motion is not changed. The bound polaron would contribute to the displacement of charge carrier in low frequency region. This is affected by the applied field.

References