

Capacitance Studies of  $\text{Nd}_2\text{CuO}_4$ <sup>†</sup>

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We have studied the dielectric properties of the  $\text{Nd}_2\text{CuO}_4$  samples that were annealed under different conditions by means of capacitance  $C(T, \omega)$  and dissipation factor  $D(T, \omega)$  measurements with the test frequency  $\omega$  in the range 20 Hz to 1 MHz and at temperature  $T$  between 5 K and 325 K. We observed two frequency-dependent peaks in the  $D(T, \omega)$  curves and corresponding features in the  $C(T, \omega)$  curves. The first peak occurs at  $T \approx 230$  K is due to the occurrence of the antiferromagnetic transition in the sample and the second one occurs at  $T \approx 120$  K is related to the change of the conduction mechanism in this system.

PACS. 77.22.Gm - Dielectric loss and relaxation.

PACS. 77.84.Bw - Elements, oxides, nitrides, borides, carbides, chalcogenides, etc.

Recent discovery of the occurrence of the electron-doped superconductivity in the  $\text{R}_{2-x}\text{Ce}_x\text{CuO}_{4-y}$  ( $\text{R} = \text{Pr}, \text{Nd}, \text{Sm}, \text{and Eu}$ ) [1,2] and  $\text{R}_{2-x}\text{Th}_x\text{CuO}_{4-y}$  ( $\text{R} = \text{Pr}, \text{Nd}, \text{and Sm}$ ) [2-4] compounds with transition temperatures up to 24 K has attracted a lot of research interest. The above compounds crystallize in a  $\text{T}'$ -phase structure in which copper is surrounded by four oxygen [5] atoms instead of the  $\text{CuO}_6$  octahedra as in the  $\text{T}$ -phase  $\text{La}_2\text{CuO}_4$  compound [6]. Recently, measurement of the ac conductivity and dielectric constant of the  $\text{T}$ -phase  $\text{La}_2\text{CuO}_4$  single crystals have been reported by different groups in the frequency ranges of 3 - 90 GHz [7] and 100 Hz - 1 MHz [8] and interesting results have been observed. Although in  $\text{La}_2\text{CuO}_4$  the  $\text{CuO}_2$  layers are under the tension of the adjacent  $\text{LaO}$  planes and can be doped with hole which creates the charge carriers in the  $\text{CuO}_2$  sheets while in the  $\text{T}'$ -phase the  $\text{CuO}_2$  planes are under compression therefore allows the electron doping. But the superconducting and normal properties are very sensitive to the doping concentration in both systems. Therefore it is interesting to compare the dielectric behaviors in both systems. In this paper, we report the dielectric properties of the  $\text{Nd}_2\text{CuO}_{4-y}$  samples by means of capacitance  $C(T, \omega)$  measurements with the test frequency  $\omega$  in the range 20 Hz to 1 MHz and at temperature between 5 K and 325 K.

The samples used in this study were prepared by solid-state reaction from high purity  $\text{Nd}_2\text{O}_3$  and  $\text{CuO}$  powders. Stoichiometric mixtures of these powder were grounded thoroughly and heated in air at 900 °C for 1 day and furnace cooled to room temperature.

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The resulting powders were pressed into pellets and fired in air at 900 °C for another day and cooled to room temperature. The pellets were then sintered in flowing He at 920 °C and 970 °C, respectively, for 1 day and cooled to room temperature at a decreasing rate of 5 °C/min. The capacitance  $C(T, \omega)$  measurements were performed on the disk-shaped pellets with silver paint coated on two sides of the pellets as the electrodes using a HP 4284A LCR meter in a He-dewar with a Si-diode as the thermometer. The radius  $R$  of the pellets is 0.5 cm and the thickness  $d$  is of 0.1 cm.

The capacitance  $C$  as a function of temperature  $T$  curves for selected test frequencies  $\omega$  for the sample annealed at 970 °C are plotted in Fig. 1. At low temperatures, up to about 100 K, the rate of increase is small and less frequency dependent. At higher temperatures the rate of increase is considerably greater and frequency dependent. The  $C(T, \omega)$  curves show distinct behaviors in different temperature regions. For  $T \leq 100$  K,  $C(T, \omega)$  decreases monotonically with decreasing  $T$  and reaches a minimum at  $T \approx 20$  K and starts to increase below this temperature. This minimum shifts to higher temperature as  $\omega$  increases and reaches a value of  $\approx 40$  K for  $\omega = 1$  MHz. Incidentally, this temperature is close to the superconducting transition temperature  $T_c$  as this compound is electron doped. For  $T \geq 100$  K,  $C(T, \omega)$  starts to increase rapidly and the value of  $C$  above a temperature  $T_0$  increases exponentially through the relation  $C(T, \omega) = a(\omega)e^{-H_1/T}$  with an activation energy of  $\approx 1520$  K (This can be seen clearly in the  $\ln C$  vs  $1000/T$  plot as shown in Fig. 2). Although  $T_0$  shifts to higher temperatures with increasing  $\omega$ , the value of  $H_1$  essentially remains the same. At  $T \geq 200$  K,  $C$  still increases with increasing  $T$ , but not with an exponential relation. Moreover, a broad bump near 250 K exists in the  $C(T, \omega)$  curve for  $\omega = 500$  Hz. near the temperature  $T_N$  where antiferromagnetic ordering occurs in this sample. With increasing  $\omega$ , the temperature for this bump increases. For the sample annealed at 920 °C, the  $C(T, \omega)$  curves exhibit similar behaviors and will not be shown here.

The logarithm of capacitance  $\ln C$  versus inverse temperature  $1000/T$  for selected test frequencies  $\omega$  were plotted in Fig. 2. A linear relation of  $\ln C$  vs  $1/T$  can be observed in the temperature between 100 K and 200 K (which depends on the test frequency  $\omega$ ). From the slope of  $\ln C$  vs  $1/T$  curves, we can deduce the value of the activation energy  $H_1 \approx 1320$  K for this compound as mentioned before. At higher temperatures, features in the  $\ln C$  vs  $1/T$  curves corresponding to the temperatures where the broad bump occurs in the  $C(T, \omega)$  curves can also be observed.

Shown in Fig. 3 are the dissipation factor  $D$  vs temperature  $T$  curves for selected values of  $\omega$  for the sample annealed at 970 °C. Two large frequency dependent peaks are observed. The first one is in the temperature range of  $\approx 250$  K (which corresponds to the temperature  $T_N$  where antiferromagnetic ordering occurs in this sample) and the other one is in the vicinity of 120 K. There is no clear feature in the  $D(T, \omega)$  curves at  $\approx 20$  K where the minima were observed in the  $C(T, \omega)$  curves. The peaks in the  $D(T, \omega)$  curves observed in this sample are similar to that of many semiconductor oxides and ferrites and has been successfully explained by the Maxwell-Wagner interfacial polarization arising from heterogeneous in the oxides such as the occurrence of the oxygen rich layers on the surface of the particles or the occurrence of the impurity ions on the grain boundaries. The low temperature peak (120 K) observed here is closely related to the occurrence of the

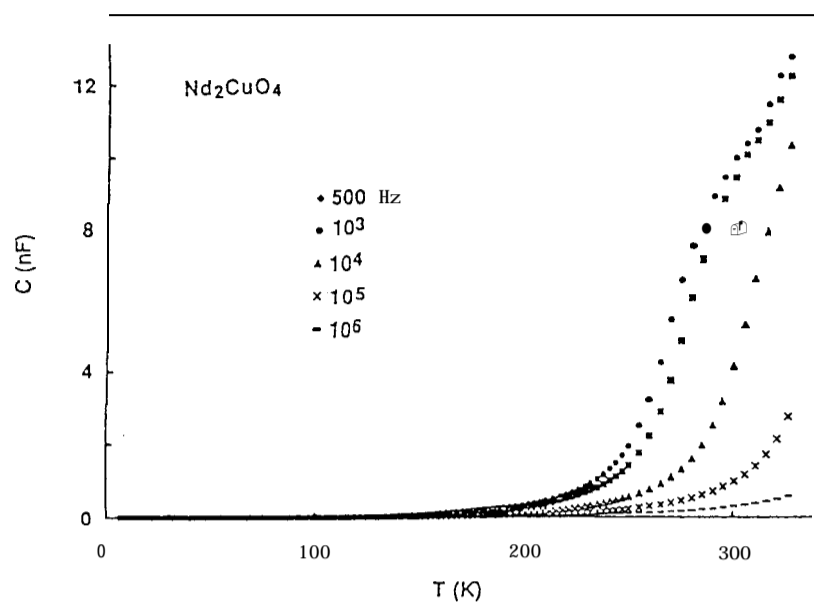


FIG. 1. The capacitance  $C$  as a function of temperature  $T$  curves for selected test frequencies  $\omega$  for the  $\text{Nd}_2\text{CuO}_{4-y}$  sample annealed at  $970^\circ\text{C}$ .

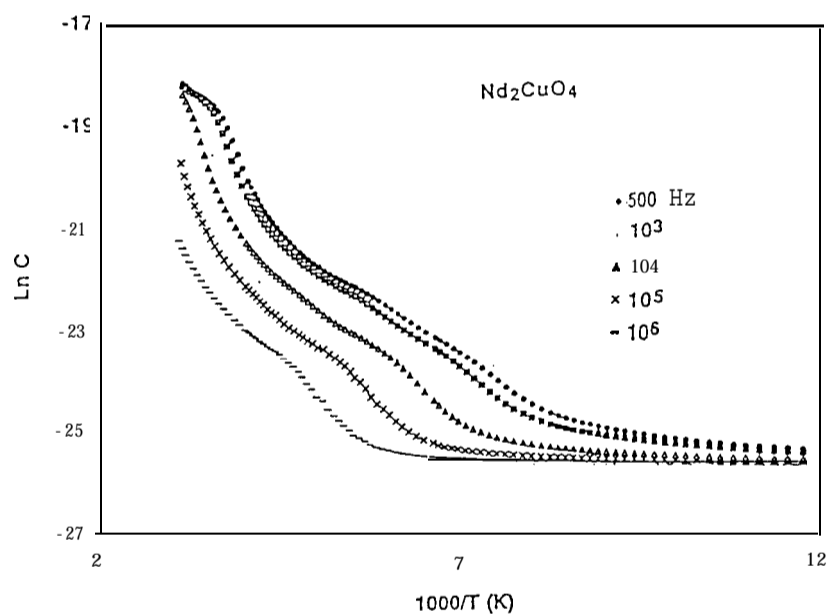


FIG. 2. The logarithm of capacitance  $\text{Ln } C$  versus inverse temperature  $1000/T$  for  $\text{Nd}_2\text{CuO}_{4-y}$  with selected test frequencies  $\omega$ .

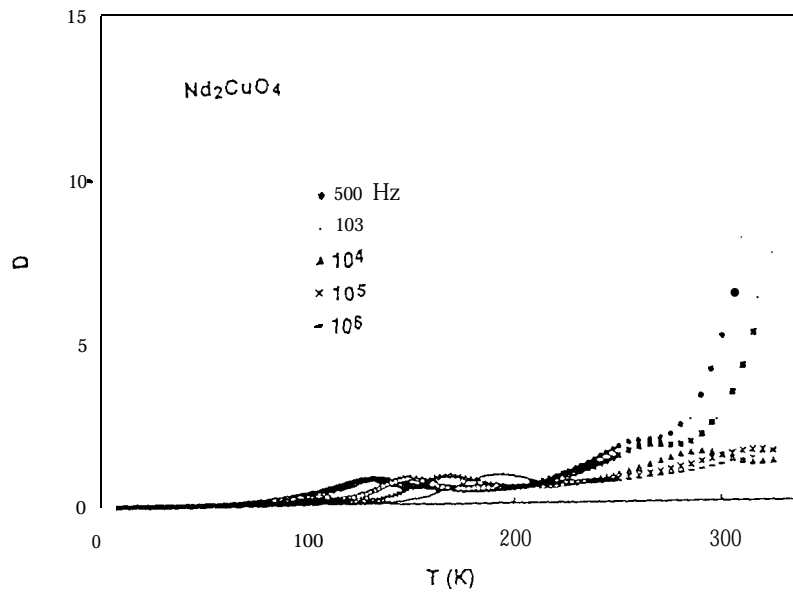


FIG. 3. The dissipation factor  $D$  vs temperature  $T$  curves for selected values of  $\omega$  for the  $\text{Nd}_2\text{CuO}_{4-y}$  sample annealed at  $970^\circ\text{C}$ .

exponential behavior in the  $C(T, \omega)$  curves. Ac conductivity data reveal that there is a change of the conduction mechanism around this temperature [9]. Although this kind of behavior has not been observed in this system before, earlier reports on the resistivity and thermal properties studies do reveal a change of the conduction mechanism near 100 K in the  $\text{La}_2\text{CuO}_4$  system [10].

In summary, we have performed the capacitance measurements on the  $\text{Nd}_2\text{CuO}_{4-y}$  samples annealed under different conditions at various test frequencies. We observed two frequency-dependent peaks in the  $D(T, \omega)$  curves and corresponding features in the  $C(T, \omega)$  curves. The first peak occurs at  $T \approx 250$  K is due to the occurrence of the antiferromagnetic transition in the sample and the second one is related to the change of the conduction mechanism in this system. This work is supported by the R.O.C. National Science Council under Grant No. NSC84-2112-M-002-018.

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