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ELECTRICAL AND MAGNETIC PROPERTIES

Anisotropy of Magnetic Properties and the Permittivity of $Nd_{1.9}Ce_{0.1}CuO_{4+\delta}$ Single Crystal

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Abstract—Dielectric and magnetic properties of the $Nd_{2-x}Ce_xCuO_{4+\delta}$ bulk single crystal with $x = 0.1$ are studied in this work. The anisotropy of the field and temperature dependences of the specific magnetization is found. The value of specific magnetization measured in the magnetic field applied in parallel to CuO_2 planes is higher than that measured in the magnetic field applied perpendicular to the planes; this is related to the additional contribution of magnetic moments of neodymium ions. In this case, at temperatures $T < 100$ K, when the magnetic field H is perpendicular to CuO_2 planes, an area of antiferromagnetic coupling of ions exists within the CuO_2 planes, which is not observed in the case of the parallel orientation of the magnetic field with respect to the CuO_2 planes. Microwave studies show the existence of strong permittivity dispersion, which indicates the presence of natural resonance frequency, whose value is beyond the frequency range of performed measurements.

Keywords: anisotropy, permittivity, magnetic moment

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INTRODUCTION

The $Nd_{2-x}Ce_xCuO_{4+\delta}$ compound is a superconductor with the electronic conductivity, which has a body-centered crystal lattice and corresponds to the tetragonal T' phase. This superconductor is characterized by a single CuO_2 plane per unit cell and does not have copper chains and apex oxygen atoms between adjacent conducting planes. Because of this, $Nd_{2-x}Ce_xCuO_{4+\delta}$ exhibits pronounced two-dimensional properties [1, 2].

The pure Nd_2CuO_4 compound is a dielectric, and the superconductivity appears only for $Nd_{2-x}Ce_xCuO_{4+\delta}$ ($0.135 \le x \le 0.20$) solid solutions based on the compound additionally annealed in an oxygen-free atmosphere; in the case of optimum alloying, the temperature of superconducting transition is $T_c \sim 24$ K at $x = 0.145$ [2–5].

It is well known that the electronic structure of high-temperature copper oxide superconductors is characterized by strong anisotropy that usually manifests itself in the temperature dependence of resistivity, which is typical of a metal and a semiconductor in measuring along the *ab* plane and *c* axis, respectively. In the case of optical measurements in the normal state along the *c* axis, the absorption edge is observed in the infrared region in contrast to metal-like spectra observed along the *ab* plane. In the superconducting state, along the *c* axis, the abrupt reflection edge is observed in the far-infrared region. The reflection edge is considered as the plasma edge of condensed carriers in the superconducting state, whereas the edge frequency is determined in crossing the frequency dependence of the real part of permittivity and zero [6].

Previous microwave experiments performed for $Nd_{2-x}Ce_xCuO_{4+\delta}$ in the CuO_2 plane were found to comply with the standard Bardeen–Cooper–Schrieffer predictions for *s* waves [7]. However, recent data that include the flux quantization [8] and penetration depth for microwave [9] and high-frequency [10] ranges have provided reliable experimental evidence of *d*-wave symmetry of pairing state [11].

In contrast to a number of experiments performed in the *ab* plane, there is little information about the properties of $Nd_{2-x}Ce_xCuO_{4+\delta}$ along the *c* axis, first of all, because of the small sizes of samples along this direction. The majority of similar experiments [5] were performed for ceramic $Nd_{2-x}Ce_xCuO_{4+\delta}$ samples. As an alternative, the reflection grazing incidence method is known to be a powerful procedure used for the determination of properties of highly anisotropic superconductors along the *c* axis [11, 12].

The magnetic properties of high-temperature superconductors with electronic conductivity have been widely studied because it was assumed that the antiferromagnetic coupling of Cu ions in layers plays

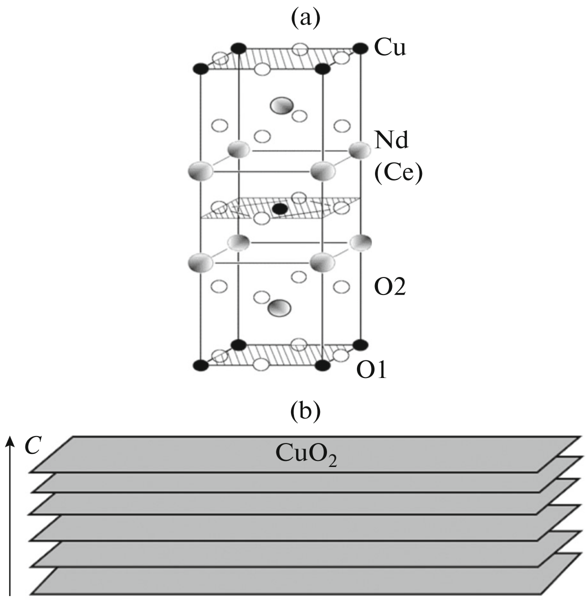

Fig. 1. (a) Structure and (b) schematic diagram of sample.

an important role in the formation of a superconducting state [13, 14].

In the previous work [15], we studied the anisotropy of the specific magnetization and magnetic susceptibility of the $Nd_{1.82}Ce_{0.18}CuO_4$ single-crystal films with different orientations of the *c* axis, which were subjected to optimum annealing. Strong anisotropic behavior of the magnetic susceptibility χ (H) at $H < 10$ kOe was found, which was related to antiferromagnetic fluctuations and the order parameter fluctuations. The highly anisotropic temperature dependence of the magnetic susceptibility also exists in the weak magnetic field $H = 100$ Oe and is related to different magnetic ordering of rare-earth magnetic ions $Nd^{3+}(Ce^{4+})$ at different orientations of the external magnetic field with respect to conducting CuO_2 planes. In this case, the presence of residual antiferromagnetic ordering of copper ions, even for the overalloying range ($x > 0.15$), leads to the magnetic anisotropy in the conducting planes. At $T < 30$ K, fluctuations of antiferromagnetic ordering lead to a decrease in the magnetic susceptibility along CuO_2 planes; this can indicate the existence of antiferromagnetic spin fluctuations slightly above the temperature of superconducting transition and appearance of the order parameter fluctuations in the CuO_2 conducting planes in the temperature range $T \sim 10–30$ K. An additional contribution of magnetic moments of Nd ions to the resulting magnetic moment was observed, particularly in the low-temperature range [15].

A complex investigation is performed in the current study of the magnetic and microwave dielectric properties of nonsuperconducting $Nd_{2-x}Ce_xCuO_{4+\delta}$ optimally annealed ($\delta \to 0$) compound with a cerium content of 10% in the CuO_2 plane and between CuO_2 planes to obtain data on the anisotropic behavior of magnetic and dielectric characteristics in the range of quantum antiferromagnet-superconductor phase transition.

EXPERIMENTAL

Bulk $Nd_{2-x}Ce_xCuO_{4+\delta}$ single crystals with $x = 0.1$ were grown from the melt using a Leonyuk ceramic crucible (MSU); the single crystals are plates 0.4 mm in thickness (d), 4.5 mm in length (a), and 4.0 mm in width (b). The orientation of the single crystal was determined using an URS-55 diffractometer. The bulk $Nd_{1.9}Ce_{0.1}CuO_4$ single crystals have a body-centered crystal lattice that corresponds to the T phase (Fig. 1a). Oxygen (O) ions are shifted from the apex positions to sites at faces of tetragonal cell; thus, the structure of the compound can be schematically represented in the form of a set of CuO_2 conducting planes separated by nonconducting buffer Nd(Ce)O layers (Fig. 1b).

Magnetic measurements were performed on a MPMS-5XL setup. Magnetization curves at several temperatures and the temperature dependence of the magnetic moment were measured. The magnetic moment was determined for two magnetic field directions, namely, parallel and perpendicular to CuO_2 planes.

The temperature dependences of the magnetic moment were measured in the temperature range $T = 4.2–300$ K after preliminary cooling of a sample to 4.2 K in the absence of external magnetic field.

To measure the field dependence of the magnetic moment, the sample was cooled to a required temperature; after that, an external magnetic field of from –30 kOe to +30 kOe was applied.

Microwave spectroscopic studies were performed at room temperature in a frequency range of from 53 to 78 GHz in accordance with the procedure described in [16]. The sample is placed in the transverse cross section of a conventional rectangular waveguide that works at the H_{10} mode. The waveguide sizes are 3.6 × 1.8 mm; the sample thickness is $d = 0.4$ mm. Wave vector of electromagnetic wave **k** is parallel to the *c* axis, i.e., perpendicular to CuO_2 planes. Vector of microwave electric field belongs to the CuO_2 plane. The microwave magnetic field belongs to the horizontal plane that is parallel to the longer side of waveguide. Using a SWR-meter and a reflectometer, magnitudes of transmission D and reflection R coefficients and the corresponding frequency dependences were determined. The obtained magnitudes of D and R were used to calculate the complex permittivity $\varepsilon = \varepsilon' - i\varepsilon''$ [16, 17].

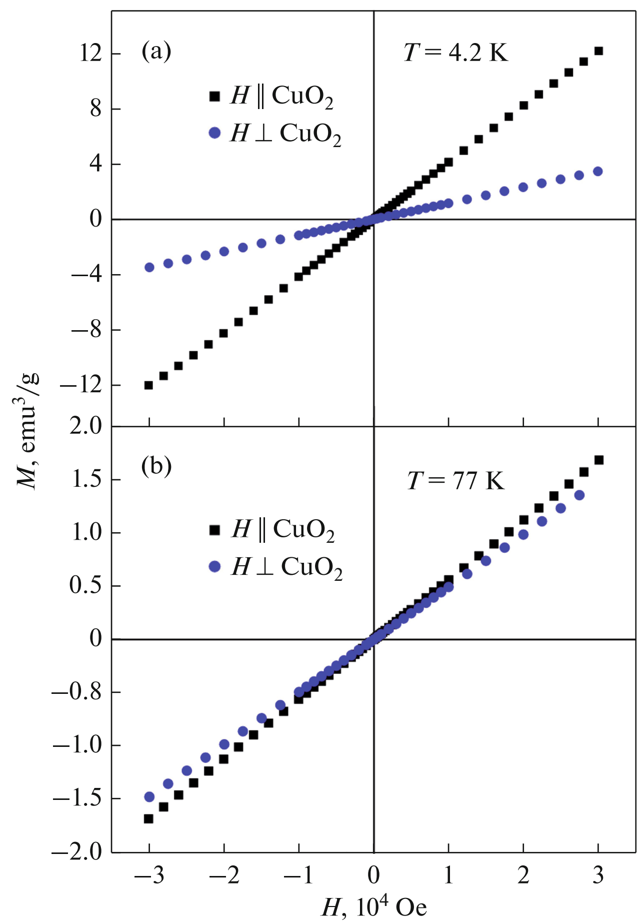

Fig. 2. Magnetization curves of the $Nd_{1.9}Ce_{0.1}CuO_{4+\delta}$ single crystal at (a) 4.2 and (b) 77 K.

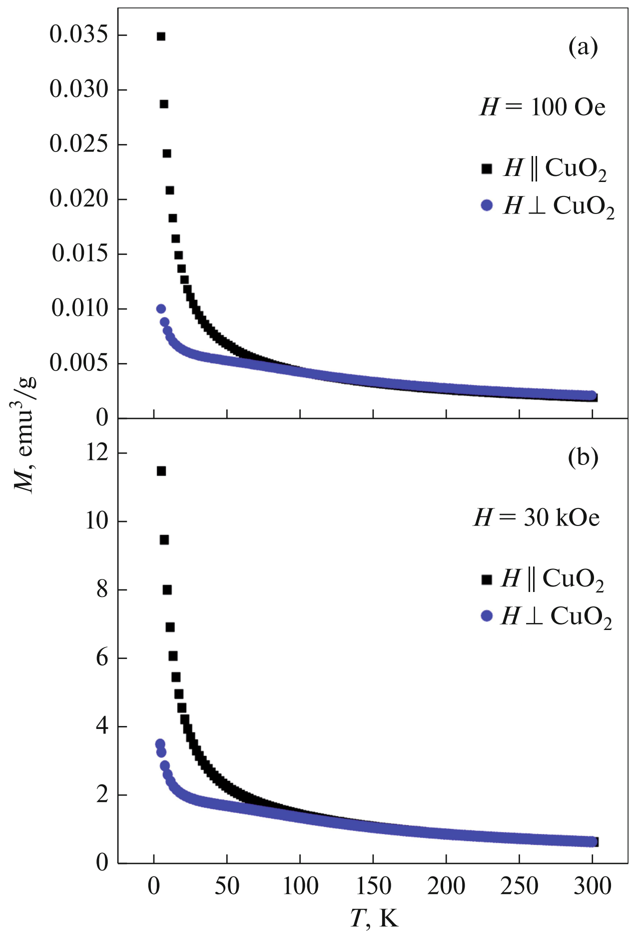

Fig. 3. Temperature dependences of the specific magnetization of the $Nd_{1.9}Ce_{0.1}CuO_{4+\delta}$ single crystal at (a) 100 Oe and (b) 30 kOe.

RESULTS AND DISCUSSION

Figure 2 shows the dependences of the specific magnetization on the magnetic field in the magnetic field range $H = -30...+30$ kOe for two fixed temperatures $T = 4.2$ K and 77 K.

As was found previously [3, 18, 19], the $Nd_{2-x}Ce_xCuO_{4+\delta}$ compound with a cerium content of 10% is not superconducting even after optimum annealing in an oxygen-free atmosphere ($\delta \to 0$); however, the compound is in close proximity to the superconducting region. Even at the cerium content $x = 0.135$, the optimally annealed compounds exhibit the superconducting transition (SC transition) at $T \sim 21.1$ K [20].

Within the magnetic field and temperature ranges under study and at two external magnetic field directions with respect to CuO_2 planes, the $Nd_{1.9}Ce_{0.1}CuO_4$ compound is found to be a paramagnet. It should be noted that the strong anisotropy of the specific magnetization of the nonsuperconducting $Nd_{1.9}Ce_{0.1}CuO_4$ compound is observed at $T = 4.2$ K and reaches $M_{\parallel}/M_{\perp} = 3.5$ in the magnetic field $H = 30$ kOe. As in [15], the specific magnetization in the external magnetic field directed in parallel to CuO_2 planes is higher than that measured along the perpendicular direction. This is related to the additional contribution of magnetic moments of Nd ions in the low-temperature range [21]. At $T = 77$ K, the specific magnetization measured in the field in parallel to CuO_2 planes decreases by an order of magnitude, namely, $M_{\parallel}$ ($T = 4.2$ K) = 12.2 emu^3/g and $M_{\parallel}$ ($T = 77$ K) = 1.5 emu^3/g, and the anisotropy disappears.

The obtained values of specific magnetization for the bulk single-crystal $Nd_{0.9}Ce_{0.1}CuO_4$ sample are $\sim 10^3$ times higher than that for the $Nd_{2-x}Ce_xCuO_{4+\delta}$ ($0 \le x \le 0.18$) single-crystal films [2, 15, 19].

The temperature dependences of the specific magnetization were measured in the external magnetic fields $H = 100$ Oe and $H = 30$ kOe (Fig. 3).

In the temperature range 100 K $\le T \le$ 300 K and for two directions of magnetic field with respect to CuO_2 planes ($H_{\parallel}$ and $H_{\perp}$), as the temperature decreases, the

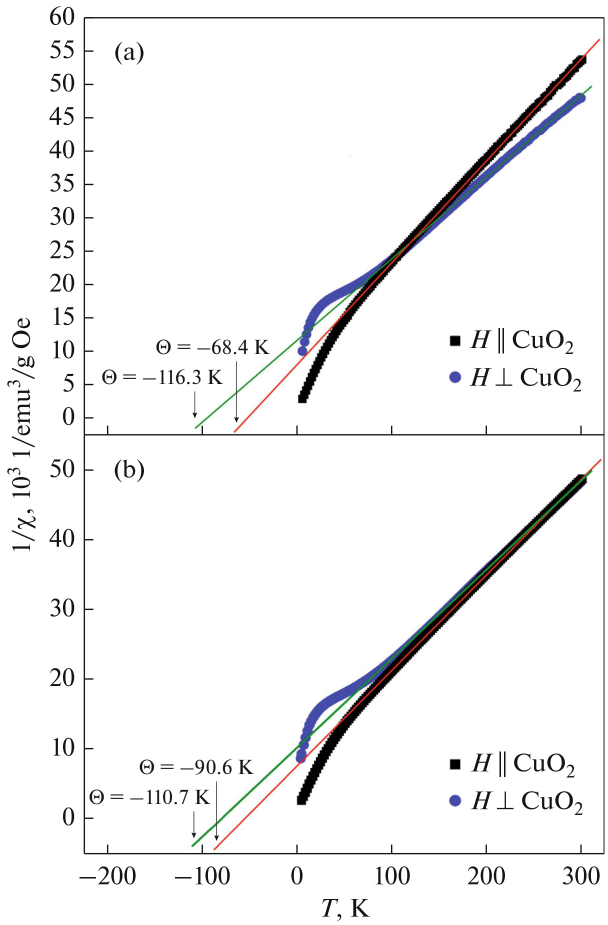

Fig. 4. Temperature dependences of the inverse magnetic susceptibility of the $Nd_{1.9}Ce_{0.1}CuO_{4+\delta}$ single crystal at (a) 100 Oe and (b) 30 kOe.

magnetic susceptibility increases in accordance with the Curie–Weiss law for paramagnets (Fig. 4):

$$\chi = \chi_0 + \frac{C}{T - \Theta}, \quad (1)$$

where χ_0 is the susceptibility at $T = 0$ extrapolated from the high-temperature range; C is the Curie–Weiss constant $C = p_{\text{eff}}^2/2.8284$, where p_{eff}^2 is the effective magnetic moment per Nd^{3+} ion; and Θ is the paramagnetic Curie temperature [22]. In the case of the magnetic field directed perpendicular to the CuO_2 planes, the important peculiarity of the behavior of $\chi(T)$ is the some deviation (decelerated increase in the susceptibility with decreasing temperature) from the Curie–Weiss law in the temperature range $T \sim$ 10–100 K. As in [23], the slight increase in $1/\chi(T)$ (in a temperature range of 30–50 K) is observed for the $Nd_{2-x}Ce_xCuO_4$ single-crystals with $x = 0$ and $x = 0.16$. This peculiarity is absent when the applied magnetic field is parallel to CuO_2 planes. Such a stabilization of the magnitude of magnetic susceptibility $\chi \approx 5 \times 10^{-5}$ emu^3/g Oe within a sufficiently wide temperature range indicates the appearance of antiferromagnetic correlations in CuO_2 planes at the field H direction perpendicular to CuO_2 planes (Fig. 4).

Table 1. Curie constant C and paramagnetic Curie temperature Θ of the $Nd_{1.9}Ce_{0.1}CuO_4$ single crystal for two directions of the magnetic field with respect to CuO_2 planes

Samples	C, K emu/g Oe	Θ, K
$H_\parallel$ 100 Oe	7.4×10^{-3}	–68.4
$H_\perp$ 100 Oe	9.7×10^{-3}	–116.3
$H_\parallel$ 30 kOe	9.8×10^{-2}	–90.6
$H_\perp$ 30 kOe	10.2×10^{-2}	–110.7

The approximation of high-temperature linear portions of the dependences 1/χ(T) allowed us to determine the Curie constant C and paramagnetic Curie temperature Θ for two configurations in the magnetic fields H = 100 Oe and H = 30 kOe (Table 1). For all samples, the parameter Θ is negative; this corresponds to the possibility of antiferromagnetic ordering.

The behavior of the dependence 1/χ(T) measured for single crystals in field H parallel to CuO_2 planes is identical to that calculated for an isolated Nd^+ ion under the effect of cubic crystalline field (in assuming that levels resulting from splitting the lower multiplet level $^4I_{9/2}$ are thermally populated and that the exchange coupling of Nd^+ ions is insignificant [24]). The anisotropy of the temperature dependence of the inverse susceptibility of both alloyed and nonalloyed crystals can result from the anisotropy of magnetic field splitting (i.e., from the separation of energy levels, which arises because of the applied magnetic field and is a function of its direction) [23]. The almost complete absence of anisotropy at high temperatures (T > 150 K) indicates a low value of the parameter B_2^0 of crystal field Hamiltonian for the tetragonal point symmetry [25].

Figure 5 shows frequency dependences of the transmission, reflection, and dissipation coefficients for the $Nd_{1.9}Ce_{0.1}CuO_{4+\delta}$ samples measured at T = 300 K. Oscillation changes in the experimental dependence of the reflection coefficient are caused by incomplete agreement of microwave tract components. Based on the data, the real and imaginary parts of the permittivity (Fig. 6) were calculated by formulas available in [16]. The average values of the parameters over the whole frequency range are ε' = 315, ε" = 53, and σ = 188 S. The obtained values of conductivity correspond to a “bad” metal. This is indicated by the high value of reflection coefficient (Fig. 5) and the sig-

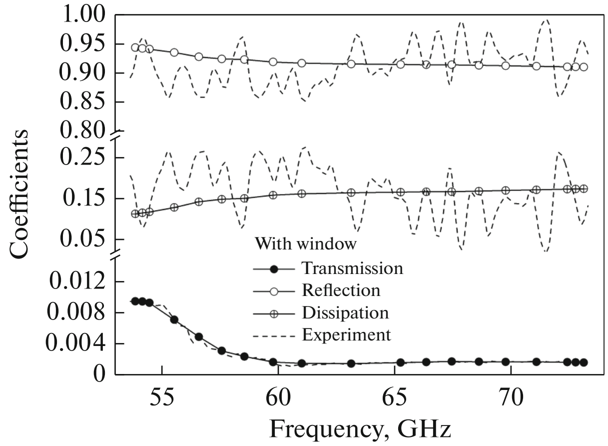

Fig. 5. Frequency dependences of the transmission, reflection, and dissipation coefficients of the $Nd_{1.9}Ce_{0.1}CuO_{4+\delta}$ single crystal.

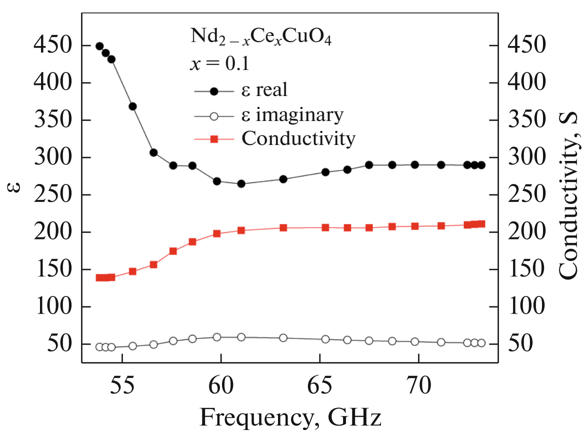

Fig. 6. Frequency dependences of the real and imaginary parts of permittivity and conductivity of the $Nd_{1.9}Ce_{0.1}CuO_{4+\delta}$ single crystal.

nificant dissipation ~0.1–0.2, i.e., the fraction of absorbed power (Fig. 6).

It was found that the strong dispersion of permittivity occurs at frequencies of 53–60 GHz; this is likely to indicate the existence of natural resonance frequency, whose value is beyond the frequency range of performed measurements. These results agree with our earlier measurements of the specific resistivity of the incompletely alloyed $Nd_{2-x}Ce_xCuO_{4+\delta}$ $(0 < x < 0.14)$ compounds, for which the nonmetallic behavior of the resistivity is observed with changing temperature ($d\rho/dT < 0$) both in conducting CuO_2 planes and in the perpendicular direction (along the c axis) [4].

CONCLUSIONS

The dielectric and magnetic properties of the bulk $Nd_{1.9}Ce_{0.1}CuO_{4+\delta}$ single crystal were studied in the range of quantum antiferromagnetic-superconductor phase transition.

It was found that the specific magnetization at $T = 4.2$ K in the external magnetic field directed in parallel to CuO_2 planes is higher than that in the perpendicular direction (along the c axis of the crystal); this is related to the additional contribution of magnetic moments of Nd ions.

It was found that, at $T < 100$ K, the antiferromagnetic coupling of ions exists in magnetic field H perpendicular to CuO_2 planes. This is confirmed by the negative value of the paramagnetic Curie temperature. In the magnetic field directed along CuO_2 planes, the transition to the antiferromagnetic coupling of ions was not found over the whole temperature range under study. The existence of strong anisotropy of the magnetization at the magnetic field directions perpendicular and parallel to CuO_2 conducting planes indicates the determining role of these planes in the appearance of quantum antiferromagnetic–superconductor phase transition as the concentration of charge carriers increases.

The microwave studies allowed us to find a 1.5 increase in the absorption coefficient at room temperature as the frequency increases from 53 to 78 GHz and the existence of strong dispersion of the permittivity, which, in our opinion, indicates the existence of natural resonance frequency, whose value is beyond the frequency range used for the measurements.

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