Incoherent interlayer transport in single-crystal films of 
Nd\textsubscript{2-x}Ce\textsubscript{x}CuO\textsubscript{4}/SrTiO\textsubscript{3}

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Abstract. The conductivity in Nd\textsubscript{2-x}Ce\textsubscript{x}CuO\textsubscript{4}/SrTiO\textsubscript{3} single-crystal films with the c-axis perpendicular or parallel to the substrate plane was studied. A comparison of the results for two types of films reveals the quasi-two-dimensional character of transport processes in stoichiometric (optimally annealed) single crystal films with 0.135 \leq x \leq 0.15 at the antiferromagnetic - superconducting phase transition.

1. Introduction

The problem of the resistivity anisotropy in the normal state of copper oxide systems has long attracted the attention of researchers. The resistivity in different directions differs not only in the magnitude but also in the character of its temperature dependence. The difference between a metallic behavior in the conducting planes (\( \rho_{ab} / dT > 0 \)) and an insulating behavior in the c-direction (\( \rho_{c} / dT < 0 \)) is a subject paid close attention by theorists [1-3], as it can give indications of the models appropriate for analyzing the normal-state resistivity of cuprates.

A strong anisotropy of the conducting properties (\( \rho_{c} / \rho_{ab} >> 1 \)) when a nonmetallic conductivity along the c axis is combined with a metallic conductivity in the ab plane was repeatedly observed in underdoped and optimally doped hole-type HTSCs [4, 5]. This is evidence of the quasi-two-dimensionality of oxide systems that consist of highly mobile CuO\textsubscript{2} layers separated by buffer layers [3]. The nonmetallic character of \( \rho_{c} \) in most superconducting high-\( T_{c} \) compounds suggests an unconventional conduction mechanism between CuO\textsubscript{2} planes.

The cerium-doped cuprate of Nd\textsubscript{2-x}Ce\textsubscript{x}CuO\textsubscript{4+δ} has a layered quasi-two-dimensional perovskite-like crystal structure [6]. As compared to other cuprate superconductors, Nd\textsubscript{2-x}Ce\textsubscript{x}CuO\textsubscript{4+δ} has many unique properties that make it an attractive subject for investigations. This is a superconductor with an electron-type conductivity whose structure contains a single CuO\textsubscript{2} plane per unit cell. In optimally annealed crystals, there are no apical oxygen atoms between neighboring conducting CuO\textsubscript{2} planes. Therefore, Nd\textsubscript{2-x}Ce\textsubscript{x}CuO\textsubscript{4+δ} crystals have clearly pronounced two-dimensional properties.

The Nd\textsubscript{2-x}Ce\textsubscript{x}CuO\textsubscript{4+δ} compound is characterized by an ability to reversibly absorb and desorb oxygen, and its properties are very sensitive to the oxygen content. Pure Nd\textsubscript{2}CuO\textsubscript{4} is dielectric, and
superconductivity appears only in related solid solutions of \( \text{Nd}_{2-x}\text{Ce}_{x}\text{CuO}_{4+\delta} \) \((x > 0.13)\) after additional annealing in an oxygen-free atmosphere \cite{7}. The main goal of such annealing is to remove excess nonstoichiometric oxygen (predominantly from the apical positions). This treatment reduces disorder, so electrons become delocalized and exhibit superconducting properties. After optimum doping and annealing, an \( \text{Nd}_{2-x}\text{Ce}_{x}\text{CuO}_4/\text{SrTiO}_3 \) crystal consists of a set of isolated conducting \( \text{CuO}_2 \) planes spaced 6 Å apart and is strongly anisotropic.

In bulk \( \text{Nd}_{2-x}\text{Ce}_{x}\text{CuO}_{4+\delta} \) single crystals, a very strong anisotropy of resistivity is observed \((\rho_c/\rho_{ab} \sim 10^4)\) \cite{8-10}. However, the nonmetallic temperature dependence of \( \rho_c(T) \) is quite rare. This is apparently due to the enhanced sensitivity of transport properties of the Nd system to the content of non-stoichiometric oxygen \((\delta)\) and difficulties in achieving an optimal annealing regime \((\delta \to 0)\) for bulk samples. On the other hand, single-crystal \( \text{Nd}_{2-x}\text{Ce}_{x}\text{CuO}_4/\text{SrTiO}_3 \) films (up to 5000 Å thick) are well suited for different annealing procedures.

Earlier we obtained and investigated high quality \( \text{Nd}_{2-x}\text{Ce}_{x}\text{CuO}_{4+\delta}/\text{SrTiO}_3 \) single-crystal films with the \( c \)-axis perpendicular \cite{11} and parallel \cite{12} to the substrate plane \((001)\) and \((1\overline{1}0)\) orientations, respectively. In \cite{13}, \( \rho_{ab}(T) \) and \( \rho_c(T) \) dependences for \( \text{Nd}_{2-x}\text{Ce}_{x}\text{CuO}_4/\text{SrTiO}_3 \) single-crystal films with \( x = 0.12 \) (nonsuperconducting underdoped films), \( x = 0.15 \) (optimally doped films with a maximal \( T_c \)) and \( x = 0.17; 0.20 \) (overdoped films) at \((001)\) and \((1\overline{1}0)\) orientations have been comparatively analyzed.

Emphasis has been placed on the overdoped region, that is, in comparison with the region of optimal doping, the "damping" region of superconductivity with a gradual decrease in \( T_c \) down to \( T_c \rightarrow 0 \) for \( x \approx 0.22 \). In our Nd-system (an \( n \)-type HTSC), we have found a transition from a quasi-2D anisotropic system \((d\rho_c/dT < 0, d\rho_{ab}/dT > 0)\) to a 3D system with metallic conductivity both in the \( ab \)-plane and in the \( c \)-axis direction \((d\rho_c/dT > 0, d\rho_{ab}/dT > 0)\) with increased doping, \( x \), and at higher temperatures, which is similar to that previously found in the La- system (a \( p \)-type HTSC) \cite{5}. A conclusion about the correlation of quasi two-dimensionality of the system with the emergence of superconductivity in copper oxide compounds can be made from \cite{5, 13}.

The region of the appearance of superconductivity with increasing cerium doping (at \( x = 0.13\pm0.14 \)) thus remained unexplored. Presently, the advances in technology have allowed us to grow high-quality \( \text{Nd}_{2-x}\text{Ce}_{x}\text{CuO}_{4+\delta}/\text{SrTiO}_3 \) single-crystal films with \( x = 0.135 \) and \( x = 0.145 \) in which the \( c \)-axis was both normal \((\{100\) films\)) and parallel to the substrate plane \((\{1\overline{1}0\) films\)) in order to study the processes of charge carrier transfer in the region of the antiferromagnetic (AFM) – superconducting (SC) quantum phase transition.

The study of the properties of these samples under optimal annealing in conjunction with the optimally doped \((x = 0.15)\) samples is the subject of this work. We are the first to experimentally observe the \( \rho_c(T) \) dependences with nonmetallic behavior for samples with \( x = 0.135 \) and 0.145 that is near the threshold \( x \) values for the AFM – SC phase transition. A comparison of the results obtained for the two types of films allowed us to demonstrate the quasi-two-dimensional character of carrier transfer in them.

2. Experimental results and discussion

In table 1, we present the characteristic parameters (film thickness and the onset and completion temperatures of the superconducting transition) for both types of the investigated \( \text{Nd}_{2-x}\text{Ce}_{x}\text{CuO}_4/\text{SrTiO}_3 \) films. The results for in-plane or out-of-plane resistivities as functions of the temperature in the samples optimally annealed in vacuum with \( x = 0.135, 0.145 \) and 0.15 are shown in figure 1 (a, b).

It is seen that the normal state conductivity in the \( ab \)-plane is metallic with the manifestation of 2D weak-localization effects at \( T \leq 70K \) in a sample with \( x = 0.135 \). The normal state out-of-plane resistivity \( \rho_c \) across the blocking layers is large with respect to the in-plane resistivity \( \rho_{ab} \) and
demonstrates a non-metallic temperature dependence \( (d\rho_c /dT < 0) \) for all the investigated films up to 300 K.

The resistivity anisotropy coefficient is large even at room temperature \( (\rho_c /\rho_{ab} \equiv 10^{-10}) \) for all the investigated films (figure 2). This parameter dramatically increases with decreasing temperature and reaches the values of \( \rho_c /\rho_{ab} \equiv 10^1 \) for the compounds with \( x=0.145 \) and 0.135 due to resistivity divergence \( \rho_c(T) \sim 1/T \) at low temperatures (see figure 1).

Table 1. The characteristic parameters for both types of investigated Nd\(_{2-x}\)Ce\(_x\)CuO\(_4\)/SrTiO\(_3\) films.

| Type of film | \( x \) | \( T_{\text{onset}}, K \) | \( T_c, K \) | \( d, \text{Å} \) |
|-------------|--------|-----------------|-----------|--------|
| (001)       | 0.135  | 13.7            | 9.6       | 3300   |
|             | 0.145  | 15.7            | 10.7      | 2500   |
|             | 0.15   | 23.5            | 22        | 2000   |
| (1\(\bar{1}\)0) | 0.135  | 12.8            | -         | 4200   |
|             | 0.145  | 15.8            | 6.3       | 4000   |
|             | 0.15   | 21.2            | 7.8       | 1400   |

Figure 1. Temperature dependencies of in-plane (left), \( \rho_{ab} \), and out-of-plane (right), \( \rho_c \), and resistivity of Nd\(_{2-x}\)Ce\(_x\)CuO\(_4\)/SrTiO\(_3\) films at different doping levels and optimal annealing for \( x = 0.135; 0.145 \) and 0.15.

Figure 2. Temperature dependence of the resistivity anisotropy coefficient for optimally annealed Nd\(_{2-x}\)Ce\(_x\)CuO\(_4\)/SrTiO\(_3\) films with various cerium contents.
A number of microscopic models for describing the deviation from a phase coherence in c-axis transport have been proposed [1-3, 14-17]. However, only the simplest one-dimensional Kronig-Penny model with its ideal periodicity (and thus the coherence) can explain the metallic nature of the interlayer conductivity.

In [14-17], the non-metallic behavior of $\rho_c(T)$ in layered oxides was attributed to incoherent tunneling of charge carriers in the c-axis direction. Incoherent transport between CuO$_2$ layers occurs when the probability of carrier scattering in the plane ($\hbar/\tau$) is much higher than the interlayer hopping integral $t_c$ ($=\hbar/\tau_{esc}$) between the planes. Here, $\tau$ is the carrier relaxation time in the plane, and $\tau_{esc}$ is the escape time from the given plane to the neighboring one.

If an electron experiences many collisions before moving to another plane, then the subsequent tunneling processes between the planes are uncorrelated. The interlayer conductivity is then proportional to the rate of tunneling between just two adjacent layers, and the diffusion coefficient along the layers ($D_\parallel$) and across them ($D_\perp$) reads (see [15], [16] and references therein):

$$D_\parallel = \frac{l^2}{2\tau}, \quad D_\perp = \frac{c^2\tau}{2\tau_{esc}}$$

where $l$ is the mean free path in the ab-plane, and $c = 6$ Å is the distance between neighboring CuO$_2$-layers. Thus, we can empirically estimate the ratio of the characteristic times as:

$$\frac{\tau_{esc}}{\tau} = (c/l)\sqrt{\rho_c/\rho_{ab}}$$

Using the estimates of the mean free path and the anisotropy coefficient ($\rho_c/\rho_{ab}$) at $T < 50$K, we get $\tau_{esc}/\tau \equiv 10$ for $x = 0.135; 0.145$ and $\tau_{esc}/\tau \equiv 2$ for $x = 0.15$. Thus, at low temperatures, the transport in the c-direction is incoherent ($\tau_{esc} \gg \tau$) for underdoped Nd systems and approaches a coherent transfer ($\tau_{esc} = \tau$) for the optimally doped one.

We emphasize that the low-temperature anisotropy coefficient is maximal for $x = 0.145$ ($\rho_c/\rho_{ab} \sim 2\cdot10^3$) and $x = 0.135$ ($\rho_c/\rho_{ab} \sim 10^3$) and much less for $x = 0.15$ ($\rho_c/\rho_{ab} \sim 10^2$) (see figure 2), in contrast to the situation in the overdoped region where the values of $\rho_c/\rho_{ab}$ and $T_c$ directly correlate (see figure 4 in [13]).

Using the model of a natural superlattice (when CuO$_2$ layers are quantum wells and Nd(Ce)O blocks serve as barriers of the effective height $\Delta$) [17-19], we can consider the disorder that is undoubtedly inherent in this system (the chaotic impurity potential) as a cause of the incoherence in the c-axis transport.

Indeed, if the wave function of the electron is localized in the c-direction with a characteristic radius of localization $r_0$, which is less than the distance between adjacent CuO$_2$ planes, then, according to [20]:

$$t_c \sim \exp(-c/r_0)\exp(-\epsilon_a/kT),$$

where $r_0^{-1} = \sqrt{2m\Delta/\hbar^2}$, and $\epsilon_a$ is the spread of electron energy in the wells due to the fluctuations of $\Delta$ values, the same as in the one-dimensional Anderson model.

The first factor in (3) (overlap integral) determines the dependence of the transition probability between the layers on the barrier height, and the second one leads to a nonmetallic temperature dependence of the conductivity at low temperatures (analogously to the conductivity within the impurity band of semiconductors [21]).

With increasing temperature, the contribution to the conductivity associated with the thermal activation of carriers across each barrier begins to play an increasingly important role [17]:
\[ \sigma_{\text{term}} \sim \exp(-\Delta/kT) \]  
(3)

and for \(kT > \Delta\), there should be a transition to "metallic" conductivity. Based on the form of the \(\rho_c(T)\) temperature dependence, we can estimate the effective barrier height as \(\Delta > 300\) K for all the investigated samples.

3. Conclusions
We have grown high-quality \((1 \overline{1} 0)\) single-crystal \(\text{Nd}_2\text{Ce}_x\text{CuO}_4/\text{SrTiO}_3\) films in which the \(c\) axis is parallel to the substrate plane, in order to exclude the effect of carrier transfer in the \(\text{CuO}_2\) planes on the transport properties along the \(c\) direction. As a result, we were able to experimentally observe nonmetallic behavior of the normal state \(\rho_c(T)\) dependence \((d\rho_c/dT < 0)\) in \(\text{Nd}_2\text{Ce}_x\text{CuO}_4\) samples both with \(x = 0.135; 0.145\) near the AFM - SC quantum phase transition and at optimal doping with \(x = 0.15\) in the SC phase.

A combination of metallic behavior of the \(\rho_{ab}(T)\) dependence and nonmetallic character of the \(\rho_c(T)\) dependence for stoichiometric (optimally annealed) underdoped and optimally doped \(\text{NdCeCuO}\) samples is an indication that the investigated system is quasi-two-dimensional. Specifically, two-dimensional conduction occurs along delocalized states in \(\text{CuO}_2\) planes, and incoherent tunneling (hopping) takes place across the blocking \(\text{Nd(Ce)}\)O layers along the \(c\) direction.

In the present work, we found that the transport in the \(c\)-direction is sufficiently incoherent in the region of coexisting antiferromagnetic and superconducting ordering at \(x = (0.13+0.14)\) and becomes more coherent in the region of SC ordering at \(x = 0.15\). We assume that the fluctuations due to competition between the AFM and SC types of ordering, as well as the impurity disorder-induced spread of electron energy levels in \(\text{CuO}_2\) quantum wells, promote the incoherent character of \(c\)-axis transport in the investigated structures.

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