

TWO-DIMENSIONAL QUANTUM INTERFERENCE CONTRIBUTIONS TO THE MAGNETORESISTANCE OF Nd_{2-x}Ce_xCuO_{4-δ} SINGLE CRYSTALS

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The 2D weak localization effects at low temperatures $T = (0.2 \div 4.2)$ K have been investigated in nonsuperconducting sample Nd_{1.88}Ce_{0.12}CuO_{4-δ} and in the normal state of the superconducting sample Nd_{1.82}Ce_{0.18}CuO_{4-δ} for $B > B_{c2} \simeq 3$ T. The phase coherence time $\tau_\varphi (\simeq 5 \cdot 10^{-11}$ s at 1.9 K) and the effective thickness of a conducting CuO₂ layer $d (\simeq 1.5 \text{ \AA})$ have been estimated by the fitting of 2D weak localization theory expressions to the magnetoresistivity data for the normal to plane and the in-plane magnetic fields. The estimation of the parameter d ensures the condition of a strong carrier confinement and makes a basis to the model of almost decoupled 2D metallic sheets for the Nd_{2-x}Ce_xCuO_{4-δ} single crystals.

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Introduction. The crystallographic structure T' of Nd_{2-x}Ce_xCuO_{4-δ} is the simplest among the superconducting cuprates, each copper atom is coordinated to four oxygen atoms in a planar structure without apical oxygen. A Nd₂CuO₄ crystal is the insulator with the valence band to be mainly of O 2*p* character and the empty conduction band to be the upper Hubbard Cu 3*d* band. The Coulomb 3*d*–3*d* repulsion on Cu site $U (\simeq 6 \div 7$ eV) is strong and it is larger than oxygen to metal charge-transfer energy $D (\simeq 1 \div 2$ eV). Thus these cuprates are classified as the charge-transfer semiconductors.

The combination of Ce doping and O reduction results in the *n*-type conduction in the CuO₂ layers. The energy band structure calculation [1] shows that the Fermi level is located in the band of *pdσ*-type formed by 3*d*($x^2 - y^2$) orbitals of Cu and *p_σ*(*x, y*) orbitals of oxygen. The *pdσ* band appears to be of highly 2D character with almost no dispersion in the normal to CuO₂ planes *z*-direction. The electrons are concentrated within the confines of conducting CuO₂ layers separated from each other by a distance $c \simeq 6 \text{ \AA}$.

Due to the layered crystal structure the high- T_c copper oxide compounds are highly anisotropic in their normal state electrical properties. The electron-doped systems Nd_{2-x}Ce_xCuO_{4-δ} exhibit a very large anisotropy factor, $\rho_c/\rho_{ab} \geq 10^4$ [2, 3] that is somewhat lower than in Bi-systems ($\rho_c/\rho_{ab} \sim 10^5$) but essentially higher than in La- and Y-systems ($\rho_c/\rho_{ab} \sim 10^2$). For the underdoped and optimally doped compounds the *c*-axis resistivity, ρ_c , is usually non-metallic ($d\rho_c/dT \leq 0$) at low enough temperatures [4]. In contrast to it, the magnitude and the temperature dependence of the resistivity in a CuO₂ plane, ρ_{ab} , are in general metallic near optimum doping.

A 2D metallic state in a system with random disorder should exhibit weak localization of the charge carriers at low temperatures [5]. Weak localization behaviour of the in-plane resistivity has been clearly observed and perfectly analysed for the Bi₂Sr₂CuO₆

systems which were investigated with high precision at T down to 0.5 K in normal and perpendicular to the CuO_2 planes magnetic fields up to 8 T [6]. As for $\text{La}_{2-x}\text{Sr}_x\text{CuO}_{4-\delta}$ or $\text{La}_{2-x}\text{Ba}_x\text{CuO}_{4-\delta}$ systems, a concentration range between the hopping regime at low x and superconducting regime at $x > 0.05$ seems to be so narrow that a well defined weak localization behaviour is difficult to observe [7, 8]. Only in the close proximity to $x = 0.05$ nonsuperconducting sample $\text{La}_{2-x}\text{Ba}_x\text{CuO}_{4-\delta}$ [7] and superconducting sample $\text{La}_{2-x}\text{Sr}_x\text{CuO}_{4-\delta}$ ($T_c = 4$ K) in the fields $B > 8$ T display some signs of weak localization ($\ln T$ -dependence of ρ_{ab}).

Due to their T' structure the Nd-systems should be particularly advantageous for the observation of 2D effects in conduction process. Really, there are several reports on the manifestation of 2D weak localization in the in-plane conductance of $\text{Nd}_{2-x}\text{Ce}_x\text{CuO}_{4-\delta}$ single crystals or films. Thus a linear dependence of resistivity on $\ln T$ comes about at $T < T_c$ for samples with $x \cong 0.15$, in which superconducting state is destroyed by a magnetic field [9]. Furthermore, a highly anisotropic (with regard to the magnetic field direction) negative magnetoresistance, predicted for 2D weak localization, has been observed in the nonsuperconducting state at low temperatures: in highly underdoped sample with $x = 0.01$ [10] and in unreduced samples with $x = 0.15$ [11] or $x = 0.18$ [12]. Measurements in superconducting $x = 0.15$ sample with high T_c ($T_c = 20$ K) has shown a similar negative magnetoresistance in high (up to 30 T) transverse magnetic fields and an upturn in the normal state resistance as T is lowered [11].

In our previous investigation of the sample with $x = 0.18$ ($T_c = 6$ K) a negative magnetoresistance has been observed after the destruction of superconductivity by a magnetic field up to 5.5 T at $T \leq 1.4$ K [13]. We report here the results of measurements at much lower temperatures (down to 0.2 K) and in the higher dc magnetic fields (up to 12 T). A drastic dependence of magnetoresistance magnitude on the direction of magnetic field is the most important experimental test for the 2D character of a conducting system. For the investigation of the magnetoresistance anisotropy we have used here the measurements on nonsuperconducting sample with x value ($x = 0.12$) which is close to the boundary value $x = 0.14$ for the superconductivity in a $\text{Nd}_{2-x}\text{Ce}_x\text{CuO}_{4-\delta}$ system.

Results. High-quality single-phase $\text{Nd}_{2-x}\text{Ce}_x\text{CuO}_{4-\delta}$ ($x = 0.12 \div 0.20$) thin films have been produced by modified laser deposition technique with a flux separation [14]. The films with thickness around 5000 Å were deposited onto hot single crystal SrTiO_2 substrate, which has (100) surface orientation. It was necessary to anneal the films subsequently in vacuum $< 10^{-2}$ torr at 800 °C during 40 min to form superconducting phase. The X-ray diffraction study has revealed the existence of the tetragonal phase only with c -axis perpendicular to the film plane. We report here the data for $\text{Nd}_{2-x}\text{Ce}_x\text{CuO}_{4-\delta}$ films with $x = 0.12$ and 0.18 only.

The in-plane resistivity ρ_{ab} and Hall coefficient R ($\mathbf{j} \parallel ab, \mathbf{B} \parallel c$) have been investigated in a single crystal superconducting film $\text{Nd}_{1.82}\text{Ce}_{0.18}\text{CuO}_{4-\delta}$ ($T_c = 6$ K) at $T = (0.2 \div 20)$ K in a magnetic field up to $B = 12$ T. In the superconducting sample the normal state transport at low T is hidden unless a magnetic field B higher than the second critical field B_{c2} is applied (for $B \perp ab$ $B_{c2} \cong 3$ T at $T = 4.2$ K). We have destroyed superconductivity by a magnetic field perpendicular to CuO_2 planes and observed a negative magnetoresistance in fields higher than B_{c2} (Fig.1) with logarithmic temperature dependence of the resistivity at $T < 4.5$ K (Fig.2). Fig.3 shows the results of measurements of the in-plane conductivity in non-superconducting sample $\text{Nd}_{1.88}\text{Ce}_{0.12}\text{CuO}_{4-\delta}$ for perpendicular $B \perp$

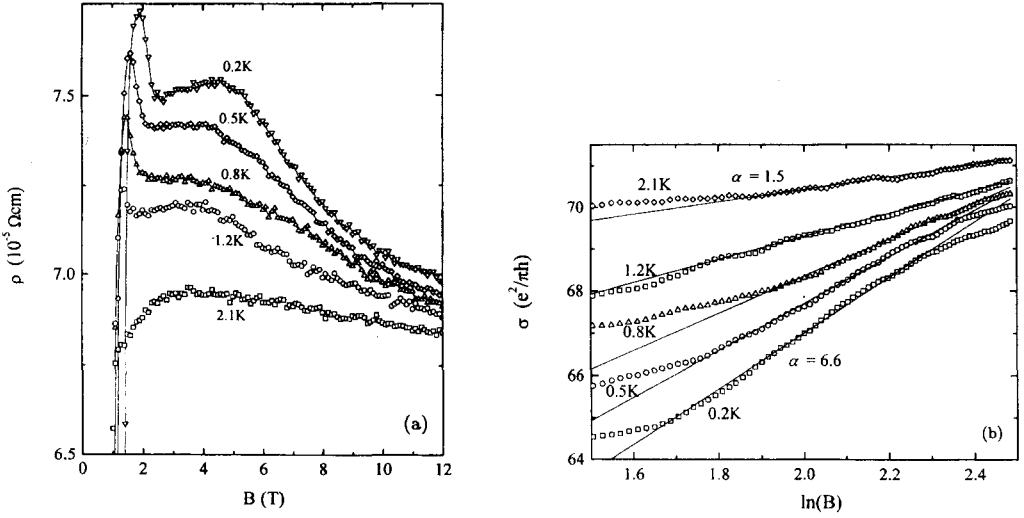


Fig.1(a). The resistivity as a function of magnetic field at different temperatures for sample with $x = 0.18$. (b). The surface conductivity as a function of $\ln B$ for sample with $x = 0.18$

and parallel B_{\parallel} to the CuO_2 planes magnetic fields up to $B = 5.5$ T at $T = 1.9$ K and 4.2 K.

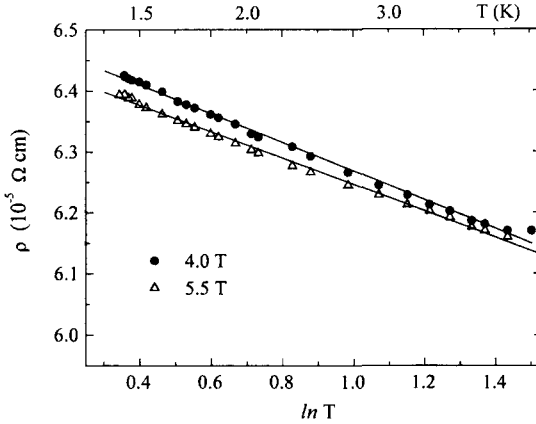


Fig.2. Logarithmic temperature dependence of the resistivity at $B > B_{c2}$ ($x = 0.18$)

Discussion. The logarithmic low temperature dependence of the conductivity is one of the indications of the interference quantum correction due to weak localization or electron-electron interaction in a 2D system. Magnetic field normal to the motion of a carrier destroys the interference leading to the localization. In 2D system it causes negative magnetoresistance for the field perpendicular to the plane but no effect for the parallel configuration. In the 2D weak localization theory the quantum correction to the Drude surface conductivity in a perpendicular magnetic field is given by [15]

$$\Delta\sigma_s(B_{\perp}) = \alpha \frac{e^2}{\pi h} \left\{ \Psi\left(\frac{1}{2} + \frac{B_{\varphi}}{B_{\perp}}\right) - \Psi\left(\frac{1}{2} + \frac{B_{tr}}{B_{\perp}}\right) \right\} \quad (1)$$

where α is a prefactor of the order of unity, Ψ is the digamma function, $B_\varphi = c\hbar/4eL_\varphi^2$ and $B_{tr} = c\hbar/2e\ell^2$. Here $L_\varphi = \sqrt{D\tau_\varphi}$ is the phase coherence length (D is the diffusion coefficient and τ_φ is the phase breaking time) and ℓ is the mean free path. At low temperature the inequality $B_\varphi \ll B_{tr}$ ($L_\varphi \gg \ell$) is valid and thus the weak localization effects is almost totally suppressed for $B \cong B_{tr}$. Let us compare the equation for the transport field, presented in the form

$$2\pi B_{tr}\ell^2 = \Phi_0 \quad (2)$$

where $\Phi_0 = \pi c\hbar/e$ is the elementary flux quantum, with the relation between the coherence length ξ and the second critical field in the so called "dirty" limit ($\xi \gg \ell$):

$$2\pi B_{c2}\ell\xi = \Phi_0. \quad (3)$$

From Eqs.(2) and (3) one has $B_{tr}/B_{c2} = \xi/\ell$ and thus the inequality $B_{tr} \gg B_{c2}$ should be valid for any dirty superconductor.

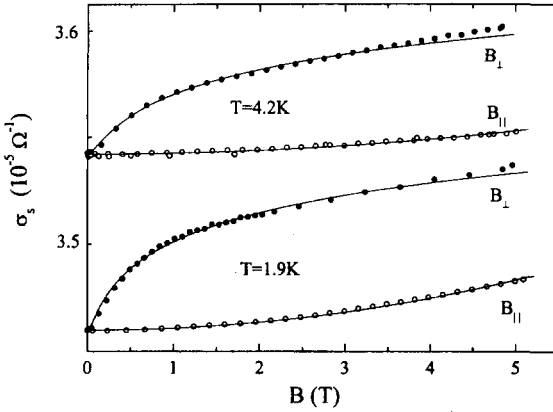


Fig.3. The surface conductivity as a function of magnetic field ($x = 0.12$)

Superconducting sample ($x = 0.18$). From the experimental values of ρ_{ab} and Hall constant R in the normal state we have obtained the Drude conductivity of a CuO_2 layer $\sigma_s = (\rho_{ab}/c)^{-1}$, the bulk $n = (eR)^{-1}$ and the surface $n_s = nc$ electron densities ($c = 6 \text{ \AA}$ is the distance between CuO_2 layers). We have $\sigma_s = 10^{-3}\Omega^{-1}$, $n = 1.1 \cdot 10^{22}\text{cm}^{-3}$ and $n_s = 6.6 \cdot 10^{14}\text{cm}^{-2}$ at $T = 4.2\text{K}$ and $B > B_{c2}$. Using the relation $\sigma_s = (e^2/h)k_F\ell$, with k_F to be the Fermi wave vector, we have estimated the parameter $k_F\ell \cong 25$. As $k_F\ell \gg 1$ a true metallic conduction in CuO_2 layers takes place.

Since $k_F = (2\pi n_s)^{1/2} \cong 6 \cdot 10^7\text{cm}^{-1}$ we have found the mean free path $\ell \cong 4 \cdot 10^{-7}\text{cm}$ and according to Eq. (2) the transport field $B_{tr} \cong 20\text{T}$. In the investigated sample the second critical field $B_{c2} \cong 3\text{T}$ at $T = 4.2\text{K}$ [13] and $B_{c2} \cong 5\text{T}$ at $T = 0.2\text{K}$. Thus we have $B_{tr} \gg B_{c2}$ and it occurs possible to observe the negative magnetoresistance owing to 2D weak localization in the interval of magnetic fields $B_{c2} < B_\perp < B_{tr}$ (Fig.1a).

In the field range $B_\varphi \ll B \ll B_{tr}$ the expression (1) may be written as

$$\Delta\sigma_s(B_\perp) = \alpha \frac{e^2}{\pi h} \left\{ -\Psi\left(\frac{1}{2}\right) - \ln \frac{B_\perp}{B_{tr}} \right\}. \quad (4)$$

Fig.1b shows the surface conductivity σ_s as a function of $\ln B$. It is seen that at $B > B_{c2}$ the experimental data can be described by simple formula (4) with prefactor α as the

only fitting parameter. At $T = 2\text{ K}$ we have $\alpha = 1.5$ but as the temperature is lowered the α value becomes essentially more than unity: $\alpha = 6.6$ at $T = 0.2\text{ K}$. Thus the effect of negative magnetoresistance at the lowest temperature is too large to be caused by the suppression of weak localization only.

In the case of effective electron attraction there exists the other orbital contribution to the negative magnetoresistance, namely, the contribution due to disorder-modified electron-electron interaction in the so called Cooper channel (the interaction of electrons with the opposite momenta) [16]. The contribution such as that may be the reason of the extra effect of negative magnetoresistance in our *in situ* superconducting sample at very low temperatures. The magnitudes of the coefficient of $\ln B$ in superconducting aluminium films at $T > T_c$ have been quantitatively explained just so [17].

When the magnetic field is applied parallel to the *ab*-plane, it turned out that the upper critical field, B_{c2}^{\parallel} , is too high and is not reached in our sample with $x = 0.18$ in a fields up to $B = 12\text{ T}$. This result is in accordance with the observation of a large anisotropy of B_{c2} for $\text{Nd}_{2-x}\text{Ce}_x\text{CuO}_{4-\delta}$ single crystals with $x = 0.16$ ($B_{c2}^{\perp} = 6.7\text{ T}$, $B_{c2}^{\parallel} = 137\text{ T}$) [18]. Thus, in order to investigate the dependence of magnetoresistance on the direction of magnetic field relative to CuO_2 plane, the study of *in situ* nonsuperconducting sample is needed.

Nonsuperconducting sample ($x = 0.12$). The positive magnetoconductivity (negative magnetoresistance) observed for this sample is obviously anisotropic relative to the direction of magnetic field (see Fig.3). From the fit of the curves $\sigma_s(B_{\perp})$ by the functional form (1) (solid curves in Fig.3) we have found the inelastic scattering length $L_{\varphi} = 550\text{ \AA}$ at $T = 1.9\text{ K}$ and $L_{\varphi} = 770\text{ \AA}$ at $T = 4.2\text{ K}$. For the in-plane diffusion coefficient $D_{\parallel} = (\pi\hbar^2/mc^2)\sigma_s$ we have $D_{\parallel} = 1.1\text{ cm}^2/\text{s}$, so that $\tau_{\varphi} = 5.4 \cdot 10^{-11}\text{ s}$ at $T = 1.9\text{ K}$ and $\tau_{\varphi} = 2.7 \cdot 10^{-11}\text{ s}$ at $T = 4.2\text{ K}$. These values are of the same order of magnitude as that obtained by Hagen et al. for $x \simeq 0.01$ crystal ($\tau_{\varphi} = 1.2 \cdot 10^{-11}\text{ s}$ at $T = 1.6\text{ K}$) [10], but in contrast to their unusual $\tau_{\varphi} \sim T^{0.4}$ dependence at $T < 10\text{ K}$ our data at $T = 1.9\text{ K}$ and $T = 4.2\text{ K}$ are compatible with the $\tau_{\varphi} \sim T^{-1}$ dependence, predicted for the electron-electron inelastic scattering in a disordered 2D system [19].

Much more weak negative magnetoresistance for parallel configuration ($B||ab$) is quadratic in B up to $B_{\parallel} = 5.5\text{ T}$ (see solid curves in Fig.3). It is of the same order of magnitude as that of Hagen et al. at $T < 5\text{ K}$ [10] or Kussmaul et al. at $T \leq 4.2\text{ K}$ [11] but we haven't seen any sign of a positive kink at $B = (1 \div 1.5)\text{ T}$ observed in [10].

Longitudinal magnetoresistance in a strictly 2D system may be caused only by the influence of the field on the spin degrees of freedom. One of the most obvious reason for negative magnetoresistance is the scattering of electrons on some spin system: the system of Cu spins or partially polarized Nd spins. For any source of spin scattering the field scale for B^2 dependence ($B \ll B_s$, $B_s = kT/g\mu_B$) is too low to explain our experimental data. For $g = 2$ $B_s = 1.5\text{ T}$ at $T = 1.9\text{ K}$ and $B_s = 3\text{ T}$ at $T = 4.2\text{ K}$, but we observe no deviations from B^2 dependence up to $B = 5.5\text{ T}$.

In standard theory of quantum interference effects in disordered conductors [20, 21] the isotropic contribution to magnetoconductivity associated with spin degrees of freedom also takes place. When the Zeeman energy of electrons $g_e\mu_B B$ exceeds kT , the magnetic field suppresses the contribution to conductivity originated from the part of electron-electron interaction thus leading to the effect of magnetoresistance. But this magnetoresistance is always positive and, with the value $g_e = 2$ for the electronic g -

factor, has the same characteristic field as that for spin scattering: $B = B_s$. Thus it apparently is not related to the effect in question, but it may be a reason of positive kink on magnetoresistance curves of Hagen et al. [10].

It is very important that in quasi-2D system with finite thickness $d \ll L_\varphi$ there exists an orbital contribution to the longitudinal magnetoresistance. It is an ordinary explanation for the parabolic negative magnetoresistance observed in parallel configuration in semiconducting 2D system: in GaAs/AlGaAs heterostructures [22] or in silicon inversion layers [23]. The finite thickness correction to the strictly 2D theory is defined by the expression [24]:

$$\Delta\sigma_s(B_{\parallel}) = \frac{e^2}{\pi h} \ln \left[1 + \left(\frac{B}{B^*} \right)^2 \right], \quad (5)$$

where $B^* = \sqrt{3}c\hbar/edL_\varphi$. It is seen from Eq.(5) that $\sigma_s(B_{\parallel})$ should be quadratic in B at fields $B \ll B^*$ with characteristic field $B^* \cong (L_\varphi/d)B_\varphi \gg B_\varphi$.

For a preliminary estimation of B^* let us assume that $d < c$ ($c = 6 \text{ \AA}$ is the distance between adjacent CuO_2 planes), then we have $B^* > 25 \text{ T}$ at $T = 1.9 \text{ K}$ and $B^* > 35 \text{ T}$ at $T = 4.2 \text{ K}$. Thus we think (so as Kussmaul et al. [11]) that the finite thickness correction to the 2D weak localization effect can reasonably explain the observed negative magnetoresistance for parallel configuration. As well as in parallel configuration there exists the finite thickness correction to the basic effect in perpendicular magnetic field (see [20], p.109). We believe that it is just the reason of the upturn of experimental points for $\sigma_s(B_{\perp})$ at $B > (3.5 \div 4) \text{ T}$ in Fig.3. By fitting of the theoretical expression (5) to the curves for $\sigma_s(B_{\parallel})$ and taking into account the values of L_φ obtained earlier, we have found for the effective thickness of a conducting CuO_2 layer $d = (1.5 \pm 0.5) \text{ \AA}$.

The value of d gives an estimate for the dimension of electron wave function in the normal to a CuO_2 plane direction and ensures the condition of a strong carrier confinement: $d < c$. It is in accordance with the proposed highly 2D character of the actual electron band of $pd\sigma$ -type with almost no dispersion along c -axis [1]. The X -ray investigations also show the concentration of electron density within the limits of $\pm 1 \text{ \AA}$ above and below a Cu atom in c -direction [25]. The single crystal NdCeCuO may therefore be regarded as multi-quantum-well system (1.5 \AA wells / 4.5 \AA barriers) or as an analog of multi-layered heterostructure. The theoretical description of high- T_c superconductors as heterostructures has been recently proposed [26].

As the 2D-version of weak localization theory is able to describe the behaviour of $\sigma_s(B, T)$ in our sample, the inequality $\tau_{esc} > \tau_\varphi$ should be valid for the escape time of electron from one CuO_2 plane to another. The escape time between adjacent quantum wells in multilayered heterostructures can be estimated from the value of the normal diffusion constant, $\tau_{esc} = c^2/D_{\perp}$. For our sample we have the anisotropy factor $D_{\parallel}/D_{\perp} \cong 10^4$ and $D_{\parallel} = 0.8 \text{ cm}^2 \cdot \text{s}^{-1}$ at 300 K . Then $\tau_{esc} \cong 4 \cdot 10^{-11} \text{ s}$ even at room temperature so that the condition $\tau_{esc} > \tau_\varphi$ may be really fulfilled at low temperatures.

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