



Doping effect on the anomalous behavior of the Hall effect in electron-doped superconductor $\text{Nd}_{2-x}\text{Ce}_x\text{CuO}_{4+\delta}$

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ABSTRACT

Transport properties of $\text{Nd}_{2-x}\text{Ce}_x\text{CuO}_{4+\delta}/\text{SrTiO}_3$ single crystal films ($B||c, J||ab$) are investigated in magnetic fields B up to $9T$ at $T = (0.4\text{--}4.2)$ K. An analysis of normal state (at $B > B_{c2}$) Hall coefficient R_H^n dependence on Ce doping takes us to a conclusion about the existence both of electron-like and hole-like contributions to transport in nominally electron-doped system. In accordance with $R_H^n(x)$ analysis an anomalous sign reversal of Hall effect in mixed state at $B < B_{c2}$ may be ascribed to a flux-flow regime for two types of carriers with opposite charges.

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1. Introduction

An observation that Hall effect in the mixed state can have a sign opposite to that in the normal state has been reported for some conventional superconductor since 1960s. The discovery of a Hall effect sign change in the most of high- T_c cuprate superconductors [1,2] stimulated a new interest to this anomalous behavior.

The high- T_c superconductors are strongly type II and thus, in accordance with the theory of Abrikosov, in the mixed state their physical properties are determined by the presence of quantized flux lines or vortices. When an external current \mathbf{J} is applied to the vortex system, the flux lines start to move under the action of the Lorentz force $\mathbf{F} = \mathbf{J} \times \mathbf{B}$ with a velocity \mathbf{v}_L . As a consequence an electric field $\mathbf{E} = -\mathbf{v}_L \times \mathbf{B}$ is induced which has resistivity component E_x and Hall component E_y , the longitudinal resistivity, $\rho_{xx} = E_x/J$ and Hall resistivity $\rho_{xy} = E_y/J$, thus appear.

Classical models which considered the hydrodynamical motion of vortices in response to a transport current in homogeneous structures (flux-flow models) were created at 1960s [3,4]. In particular, Bardeen and Stephen (BS) [3] have found that in the mixed state a part of the current flows through the vortex core that results in both dissipation and Hall effect. The core is treated as a normal conductor and thus in this model the Hall effect should have the same sign as in the normal state.

Various theoretical models have been proposed to explain the sign change of the Hall resistivity in the mixed state (for an extensive review see [5]) but the origin of this phenomenon remains controversial.

If the proper pinning forces are included in the equation of vortex motion [6,7] it leads to a back-flow current and to a possibility of Hall effect sign reversal. However, pinning forces are highly sample dependent and thus difficult to analyze.

Among the other models for the explanation of the Hall effect sign change in the mixed state a two-band model was created by Hirsch and Marsiglio (HM) [8] with electrons and holes supposed to have rather different superconducting gaps. Such a model will suggest a simple and natural explanation of the phenomenon but only if there are grounds to consider that two types of carriers, namely, electrons and holes, really coexist in the systems studied.

There are much recent activities in investigation of possible electrons and holes coexistence in the normal state of cuprate superconductors [9]. In particular, in electron-doped superconducting cuprates, angle resolved photoemission spectroscopy (ARPES) [10–13] and transport studies [9,14–21] have shown that both electrons and holes play a role in the normal state properties. Two kinds of carriers in electron-doped cuprates with different lanthanide cations seem to arise from the electronic structure near the Fermi surface (FS) of the CuO_2 planes.

ARPES studies in NdCeCuO [11,22,23] have revealed a small electron-like FS pocket in the underdoped region, and a simultaneous presence of both electron- and hole-like pockets near optimal doping. The conclusions of the ARPES measurements and

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first-principle calculations of the electronic structure on the electron-doped high- T_c superconductors $\text{Ln}_{1.85}\text{Ce}_{0.15}\text{CuO}_4$ ($\text{Ln} = \text{Nd}, \text{Sm}$ and Eu), performed by Ikeda et al. [12,13] are in accordance with the results of papers [11,22,23].

A spin density wave (SDW) model [24] was proposed which gives qualitative explanation to ARPES observations. In this model, SDW ordering would induce FS reconstruction that results in an evolution from an electron pocket to the coexistence of electron-like and hole-like pockets and then into a single hole-like FS with increasing of doping. At present the development of this theoretical model is continued [25,26].

A model of FS reconstruction, induced by SDW, was equally well used for interpretation both of high-field Hall effect and magnetoresistance in electron-doped $\text{Pr}_{2-x}\text{Ce}_x\text{CuO}_4$ films [16] and of doping dependence of Shubnikov-de-Haas (SdH) oscillations in $\text{Nd}_{2-x}\text{Ce}_x\text{CuO}_4$ single crystals [19,20]. A two-carrier model with coexisting of electrons and holes turns out to be suitable also for describing of temperature and doping dependencies of normal state Hall effect and thermopower in $\text{Nd}_{2-x}\text{Ce}_x\text{CuO}_4$ [9,14], $\text{Pr}_{2-x}\text{Ce}_x\text{CuO}_4$ [15] and $\text{La}_{1.85}\text{Ce}_{0.15}\text{CuO}_4$ thin films [17,18].

Thus, it seems to us that it is just the time to make a new attempt of describing the behavior of the mixed state Hall effect in electron-doped superconductors on the ground of a two-carrier model. In this paper we study magnetic field dependencies of longitudinal and Hall resistivities of electron-doped superconductor $\text{Nd}_{2-x}\text{Ce}_x\text{CuO}_{4+\delta}$ with various Ce content both in the normal and the mixed states.

2. Samples and equipment

The series of $\text{Nd}_{2-x}\text{Ce}_x\text{CuO}_{4+\delta}/\text{SrTiO}_3$ epitaxial films ($x = 0.14, 0.15, 0.17, 0.18$) with standard (001) orientation were synthesized by pulsed laser evaporation [21]. The original target (the sintered ceramic tablet of $\text{Nd}_{2-x}\text{Ce}_x\text{CuO}_{4+\delta}$ of the given composition) was evaporated by a focused laser beam and the evaporated target material was deposited on a heated single-crystal substrate. The substrate material was SrTiO_3 with (100) orientation and dimensions of $5 \times 10 \times 1.5$ mm. The substrate temperature was 800°C , the pressure during the deposition was 1.067 mbar, the residual gas was nitrous oxide (N_2O). Then the films were subjected to heat treatment (annealing) under various conditions to obtain samples with various oxygen content. As a result, three types of samples with $x = 0.15$ were obtained: as-grown samples, optimally reduced samples (optimally annealed in a vacuum at $T = 780^\circ\text{C}$ for $t = 60$ min; $p = 1.33 \times 10^{-2}$ mbar) and nonoptimally reduced samples (annealed in a vacuum $T = 780^\circ\text{C}$ for $t = 40$ min; $p = 1.33 \times 10^{-2}$ mbar), five types of samples with $x = 0.14$: optimally annealed in a vacuum at $T = 780^\circ\text{C}$ for $t = 25$ min; $p = 1.33 \times 10^{-2}$ mbar and four nonoptimally reduced samples (annealed in a vacuum $T = 780^\circ\text{C}$ for $t = 5, 20, 30, 64$ min; $p = 1.33 \times 10^{-2}$ mbar), ten types of samples with $x = 0.17$: optimally annealed in a vacuum at $T = 780^\circ\text{C}$ for $t = 50$ min; $p = 1.33 \times 10^{-2}$ mbar) and nonoptimally reduced samples (annealed in a vacuum $T = 780^\circ\text{C}$ for different times; $p = 1.33 \times 10^{-2}$ mbar) and five types of samples with $x = 0.18$: optimally annealed in a vacuum at $T = 600^\circ\text{C}$ for $t = 35$ min; $p = 1.33 \times 10^{-5}$ mbar and four nonoptimally reduced samples (annealed in a vacuum $T = 600^\circ\text{C}$ for $t = 10, 15, 25, 60$ min; $p = 1.33 \times 10^{-5}$ mbar) The film thickness was 1200–3800 Å. The annealing conditions and transition temperatures of optimally reduced films and experimental values of the upper critical fields are presented in Table 1.

A part of experimental results on the galvanomagnetic properties of these films are presented earlier [27]. In particular, in our works [27a] the upper critical fields B_{c2} were determined for

Table 1

Optimal annealing conditions, temperatures of the superconducting (SC) transition and experimental values of the upper critical fields at the minimal temperature of the measurements for $\text{Nd}_{2-x}\text{Ce}_x\text{CuO}_{4+\delta}/\text{SrTiO}_3$ epitaxial films with different cerium doping.

x	Annealing conditions	T_c^{onset} (K)	T_c (K)	B_{c2} (T)
0.14	$T = 780^\circ\text{C}$ $p = 1.33 \times 10^{-2}$ mbar $t = 25$ min	12.1	8.6	2.9 ($T = 1.4$ K)
0.15	$T = 780^\circ\text{C}$ $p = 1.33 \times 10^{-2}$ mbar $t = 60$ min	21.1	20.0	7.1 ($T = 0.4$ K)
0.17	$T = 780^\circ\text{C}$ $p = 1.33 \times 10^{-2}$ mbar $t = 50$ min	12.8	11.0	1.7 ($T = 3$ K)
0.18	$T = 600^\circ\text{C}$ $p = 1.33 \times 10^{-5}$ mbar $t = 25$ min	7.7	6.4	0.9 ($T = 1.6$ K)

$\text{Nd}_{1.85}\text{Ce}_{0.15}\text{CuO}_{4+\delta}$ single crystal films by so named “resistive” method: from the shift of the resistive transition into the superconducting state in an external magnetic field. For all of investigated $\text{Nd}_{1.85}\text{Ce}_{0.15}\text{CuO}_{4+\delta}$ samples an increase of the external magnetic field resulted in a real shift of the resistive transition to lower temperatures without appreciable broadening. Although superconducting fluctuations led to a rounding of the transition, it was still possible to deduce a critical field from the position of the transition by extrapolating its steep part to the normal state value. In [27a] we have presented B_{c2} (T) dependences corresponding to 0.5 ρ_n criterion.

Now we report on the investigation results of the magnetic field dependent Hall coefficient as a function of doped electron concentration in optimally reduced films. The sharp resistive transition into the superconducting state and the high critical temperature of the optimally reduced samples confirm the high quality of the films under investigation. Hall effect and magnetoresistance were measured by a standard dc technique using a 12T Oxford Instruments superconducting magnet in the temperature range $T = (0.4/4.2)$ K.

3. Experimental results and discussion

The in-plane longitudinal ρ_{xx} and Hall ρ_{xy} components of the resistivity are measured as the functions of magnetic field B perpendicular to the ab-plane up to 9T in single crystal films of electron-doped superconductor $\text{Nd}_{2-x}\text{Ce}_x\text{CuO}_{4+\delta}$ at $T = (0.4\text{--}4.2)$ K. Fig. 1 shows the field dependences of the resistivities ρ_{xx} and ρ_{xy} for optimally reduced $\text{Nd}_{2-x}\text{Ce}_x\text{CuO}_{4+\delta}$ films with $x = 0.14$ (a) and 0.15 (b) and Fig. 2 shows $\rho_{xx}(B)$ and $\rho_{xy}(B)$ dependences for optimally reduced $\text{Nd}_{2-x}\text{Ce}_x\text{CuO}_{4+\delta}$ films with $x = 0.17$ (a) and 0.18 (b) for $T = 4.2$ K. Here B_p is a vortex-depinning field and B_{c2} is the upper critical field, determined as it is shown in Fig. 1b, that, distracting the rounding of a transition due to a superconducting fluctuation, marks a field of the transition to the normal state both of ρ_{xx} and ρ_{xy} . Arguments in favor of “onset” criterion for an estimation of B_{c2} based on peculiar features of the vortex system behavior in a high-temperature superconductor are given by Gantmaher et al. [28] for $\text{Nd}_{2-x}\text{Ce}_x\text{CuO}_{4+y}$ single crystals and by Jaroszyński et al. [29] for $\text{NdFeAsO}_{0.7}\text{F}_{0.3}$ single crystals. The obtained B_{c2} values are presented in Table 1. The region $B_p < B < B_{c2}$ corresponds to a mixed (vortex) state where finite resistivity is a consequence of vortex moving under the action of the Lorentz force.

The evolution of the Hall coefficient value in the normal state above the upper critical field B_{c2} is traced with a variation of Ce

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