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Anisotropic low-temperature in-plane magnetoresistance in electron doped $Nd_{2-x}Ce_xCuO_{4+\delta}$

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Nominally electron doped antiferromagnetic tetragonal nonsuperconducting $Nd_{2-x}Ce_xCuO_{4+\delta}$ ($x = 0.12$) has been shown to manifest strong angular dependence of the in-plane magnetoresistance on the orientation of the external magnetic field within the *ab* plane in many aspects similar to that observed in hole doped $YBa_2Cu_3O_{7-\delta}$ and $La_{2-x}Sr_xCuO_4$. Specific fourfold angular magnetoresistance anisotropy amounting to several percents was observed in oxygen annealed films at low temperatures and in external magnetic field up to 5.5 T. Strong temperature dependence and fourfold symmetry observed in our sample points to a specific role of rare-earth (Nd) ions in magnetoresistance anisotropy. At low temperature $T = 1.4$ K we observed the unusual transformation of magnetoresistance response with increasing the external magnetic field which seems to be a manifestation of a combined effect of a crossover between first and second order spin-flop transitions and a field-dependent rare-earth contribution to quasiparticle magnetotransport.

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The unusual spin and charge dynamics in high-T_c cuprates have been attracting great interest. Especially, in the underdoped region of hole-doped cuprates, many anomalous features have been unveiled. Unusual magnetoresistance anomalies were reported recently for the heavily underdoped antiferromagnetic $TmBa_2Cu_3O_{6+x}$ ($x = 0.30$) by Amitin et al. [1], $YBa_2Cu_3O_{6+x}$ ($x = 0.30; 0.32$) by Y. Ando et al. [2], $YBa_2Cu_3O_{6+x}$ ($x = 0.25$) by E. Cimpoiasu et al. [3], $La_{2-x}Sr_xCuO_4$ by Y. Ando et al. [4]. The in-plane resistivity ρ_{ab} in all the crystals exhibits unconventional metal-dielectric duality with the high-temperature ($T > 50$ K) metal-like behavior in contrast with the low-T insulating one which is not compatible both with that for a simple band insulator and for an Anderson insulator. The crystals demonstrate an unusual behavior of the in-plane magnetoresistance, $\Delta\rho_{ab}/\rho_{ab}$ when the magnetic field **H** is applied along the CuO_2 planes. These are a striking $d-wave$ shaped ($\propto \cos 2\phi$) angular dependence with a clear $\propto 1/T$ temperature dependence, anomalous low-field behavior with saturation above a well-defined threshold field, and hysteretic effects at low temperatures.

Anisotropic magnetoresistance (AMR) anomalies observed in heavily underdoped hole-doped 123 and 214 cuprates are believed to be an important signature of the interplay between the charge and spin subsystems that seemingly plays a central role in the physics of high-temperature superconductors. This interplay, tuned by a charge doping, underlies the dramatic changes in the physical properties across the phase diagram of these materials. Its study may provide a valuable information as regards the nature both of charge carriers and magnetism in cuprates, their role in high-T_c, and the origin of magnetoresistance anomalies. In this connection, it is strongly desirable to extend the respective studying on other cuprates, in particular, to the electron doped ones. Indeed, the question arises, whether the features generic to hole-doped cuprates are observable in the electron doped ones? May be, these features are inherent only to hole carriers as it was argued in a microscopic model by Moskvin and Panov [5]?

The properties of the electron doped family of cuprates, have been studied to a lesser extent than those for hole doped counterparts. Just recently, Lavrov et al. [6] have shown that a magnetic-field-induced transition from noncollinear to collinear spin arrangement in adjacent CuO_2 planes of lightly electron-doped $Pr_{1-3x}La_{0.7}Ce_xCuO_4$ ($x = 0.01$) crystals affects significantly both the in-plane and out-of-plane resistivity. In the high-field collinear state, the magnetoresistance does not saturate but exhibits an intriguing fourfold-symmetric angular dependence, oscillating from being positive at **H** ∥ [100] to being negative at **H** ∥ [110] with $\Delta\rho_{ab}/\rho_{ab}$ exceeding 30% at low temperatures. Fournier et al. [7] have found sharp fourfold magnetoresistance oscillations for nonsuperconducting $Pr_{2-x}Ce_xCuO_4$ crystals at a substantially larger electron doping ($x = 0.15$). The question arises, what is the mechanism of the revealed MR features, and whether these features are observable in other electron doped cuprates?

The main goal of our paper was to investigate the low-temperature magnetoresistance in thin films of underdoped $Nd_{2-x}Ce_xCuO_{4+\delta}$ [8]. It turned out, that the oxygen annealed films indeed reveal the anisotropic magnetoresistance which angular dependence exhibits anomalous field dependence evidencing the unconventional character of first-to-second order crossover for spin-reorientation transition.

The c-axis-oriented epitaxial $Nd_{2-x}Ce_xCuO_{4+\delta}$ films with a thicknesses of 2500 Å and fixed contents of cerium $x = 0.12$ were grown using pulsed laser ablation from a ceramic target. The process consists of the evaporation of ceramic target by a focused laser beam and subsequent deposition of the target material on a heated single-crystal $SrTiO_3$ substrate with an orientation [100] (with the size of 5×10 mm and thickness of 1.5 mm); the temperature of the substrate was 800°C; the deposition pressure was 0.8 Torr; the residual gas was air; and the target was sintered ceramic pellet of $Nd_{2-x}Ce_xCuO_{4+\delta}$ of a specified composition. In the thus obtained single-crystal films, the CuO_2 (ab) plane coincided with the plane of the substrate. To obtain samples with various oxygen content, we made use of two regimes of annealing: "the optimum annealing" (60 min, $T = 780\,°C$, $p = 10^{-3}$ Torr) and "annealing in oxygen" (60 min, $T = 500\,°C$, $p = 760$ Torr). The x-ray diffraction measurements showed the $Nd_{2-x}Ce_xCuO_{4+\delta}$ single-crystalline films on $SrTiO_3$ substrates are epitaxial with [001] orientation of the surface plane and a [001] misorientation of domains less than one degree.

The resistivity measurements were performed with a standard four-probe technique and a superconducting solenoid in a dc magnetic field of up to $H = 6$ T in a temperature range of $1.4 \div 300$ K. First we should note that both as grown and annealed samples reveal the temperature dependence of resistivity [11] typical for hole-underdoped cuprates [2, 4] with high-temperature metallic and low-temperature insulating behavior. In Fig.1 we present the temperature dependence of resistivity for as grown and the oxygen annealed samples. To detect the in-plane magnetoresistance anisotropy, that is the dependence of magnetoresistance on the angle between the direction of current and that of external magnetic field, the sample was rotated around c-axis by $0 \div 270$ degrees with a fixed in-plane orientation of magnetic field. For the oxygen annealed sample we actually found the

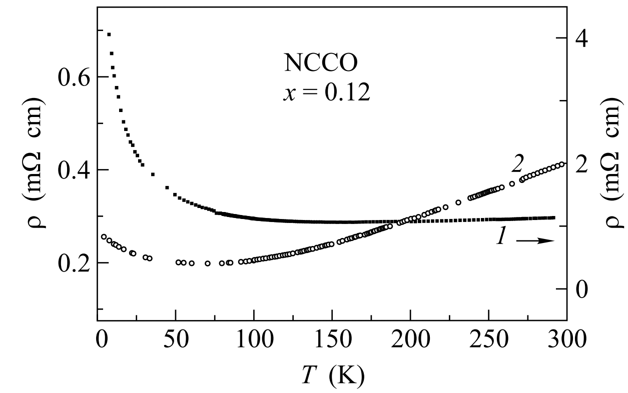

Fig.1. The temperature dependence of resistivity for the oxygen annealed (1) and "as grown" (2) thin films of $Nd_{2-x}Ce_xCuO_{4+\delta}$ ($x = 0.12$)

in-plane magnetoresistance anisotropy with minima and maxima alternating in 45° thus forming the four-lobe rose. In Fig.2 we present the raw experimental mag-

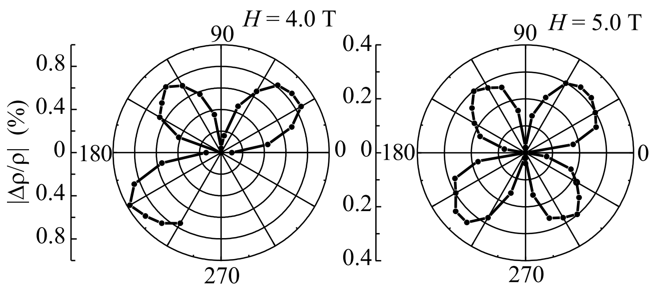

Fig.2. The angular dependence of magnetoresistance $\Delta\rho(h,\phi)/\rho(h,\phi = 0)$ in the oxygen annealed thin film $Nd_{2-x}Ce_xCuO_{4+\delta}$ ($x = 0.12$) at $T = 4.2$ K

netoresistance data obtained at $T = 4.2$ K and external magnetic field 4 and 5 T (the latter pattern was continued to the whole angle range). In both cases we clearly see the fourfold angular symmetry of the AMR effect. Detailed field behavior of AMR was undertaken at low temperature 1.4 K. The close examination of the field dependence of the AMR at $T = 1.4$ K (Fig.3) have revealed its unexpected behavior. Indeed, at relatively small external field 1.5 T we deal with a fourfold four-lobe rose with lobes oriented along $\pm\pi/4, \pm3\pi/4$. On increasing the applied field from 1.5 up to 3.5 T we arrive at a fairly visible lobes broadening firstly with a flat then with a concave top. Then, at $H_{ext} = 4.0$ T we see a puzzling effect of the lobes "splitting" accompanied by a sharp decrease of the AMR effect. Further increase of the field leads to the closing in of different half-lobes along directions $\pm\pi/2, \pm3\pi/2$, so at $H_{ext} = 4.5$ T we see a fourfold four-lobe rose rotated $\pm\pi/4$ with respect to that typical for small external fields. The effect is accompanied

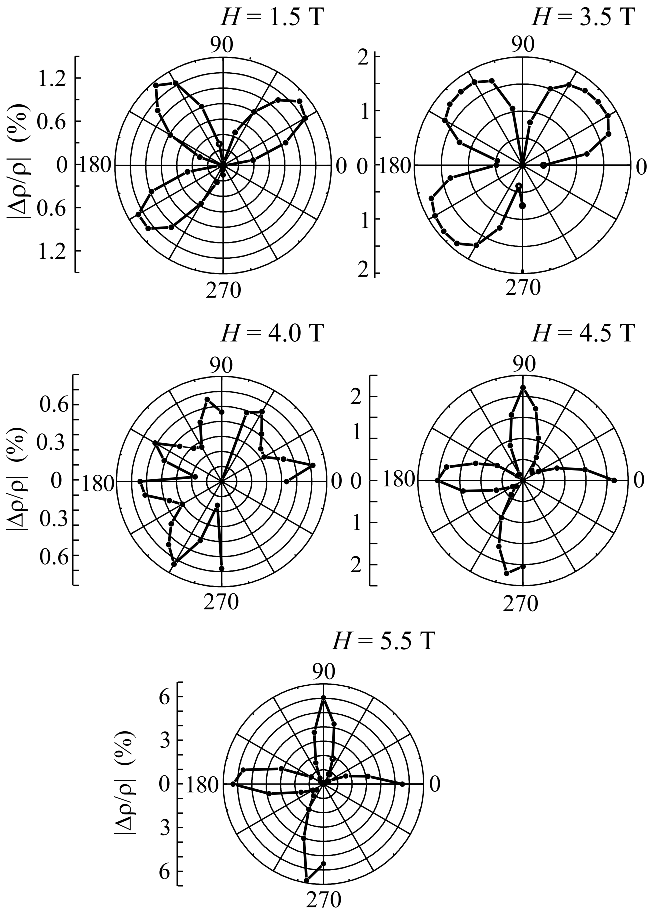

Fig.3. Transformation of AMR in NdCeCuO film at $T = 1.4$ K in a rising external field as an evidence of unconventional first-to-second order crossover spin-reorientation transition

by a strong increase of the AMR, particularly visible at $H_{\text{ext}} = 5.5$ T when it an order of magnitude exceeds the 4.2 K effect. This unconventional field behavior of AMR most likely evidences an unusual rearrangement of magnetic structure induced by an external field.

In Fig.4 we have presented the field dependence of magnetoresistivity for the field oriented along the direction of current measurement. The field effect at $T = 4.2$ K looks like that for 123 and 214 systems [2, 4]. However, the lowering the temperature to the range of Nd ordering leads to a puzzling *reentrant* effect when the magnetoresistance first falls up to $H = 3.5$ T and then rises backwards up to small values at $H = 5$ T. It is worth noting that the *reentrant* behavior of magnetoresistance reflects an interplay between isotropic and anisotropic contributions. We did not observe sizeable hysteretic effects, that likely points to a manifestation of the effects of the rotation of antiferromagnetic vector rather than the effects of the shift of domain walls.

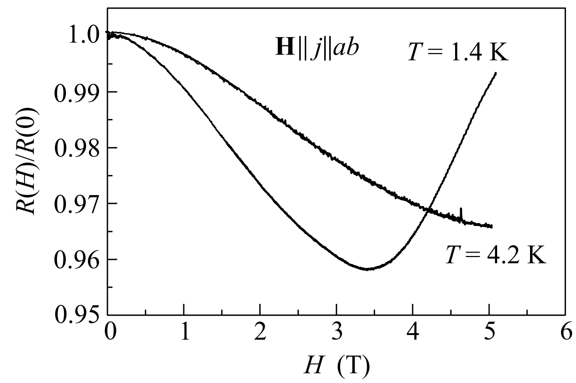

Fig.4. The field dependence of magnetoresistance in the oxygen annealed thin film of $Nd_{2-x}Ce_xCuO_{4+\delta}$ $(x = 0.12)$

Turning to the discussion of the experimental data we first would like to note that the complete interpretation of anisotropic magnetoresistance in electron/hole doped cuprates implies the knowledge of its electron structure, the structure of charge carriers, magnetic structure and its transformation in external magnetic field.

Understanding the anomalous features of the in-plane and out-of-plane normal state transport, particularly magnetotransport, in layered cuprates remains a challenge. Even in the simplest form we deal with the general problem of the transport properties of a single electron or hole in a strongly correlated antiferromagnetically ordered quasi-2D cuprate which continues to be the topic of much debate, both theoretically and experimentally.

The first purely qualitative scenario of anisotropic magnetoresistance in heavily underdoped cuprates was suggested by Ando et al. [2]. The authors consider qualitatively these features to be a manifestation of the "charge stripe" ferromagnetic structure in 123 system, which could be easily rotated by a rather small external magnetic field. The stripe scenario was first questioned by Janossy et al. [12]. In their view the anomalous magnetoresistance is due to an *ab* plane anisotropy of the resistivity in the bulk and to a magnetic field dependent antiferromagnetic (AFM) domain structure (magnetic inhomogeneity). The phenomenological symmetry based theory of anisotropic magnetoresistance in the underdoped easy-plane antiferromagnets of $YBa_2Cu_3O_{7-\delta}$-type has been developed by Gomonaj and Loktev [13] and by Cimpoiasu et al. [3]. The in-plane resistivity was assumed to be a symmetry allowed function of the components of the in-plane strain tensor, produced by the magnetoelastic coupling [3]. Gomonaj and Loktev [13] addressed the in-plane resistivity to be a symmetry allowed function of the in-plane components of antiferromagnetic vector **L**. The field and angular dependencies of the resistivity tensor in 123 system have been shown to be in a satisfactory agreement with the available experimental data for the low-field polydomain and high-field single-domain phases [3, 13]. However, despite the phenomenological approaches provide a fairly reasonable explanation of many aspects of anisotropic magnetoresistance phenomenon, they fail to explain the magnitude of the effect observed and strong $1/T$ temperature dependences both of isotropic and anisotropic magnetoresistance observed in 123 and 214 systems. Such a low-temperature anomaly is absolutely incompatible with the expected saturation of magnetic Cu subsystem. Moskvin and Panov [5] have proposed a microscopic scenario for *hole* magnetoresistance, in which the features of the in-plane anisotropic magnetoresistance are attributed to the spin-induced orbital polarization of the triplet state formed by a doped hole in the CuO_4 centers. The model is by now the only microscopic theory that seems to provide the most consistent interpretation of magnetoresistance anomalies in cuprates. The model implies a quasi-degeneracy in the ground state of the two-hole CuO_4^{5-} center with the two close in energy $^1A_{1g}$ and ^1E_u terms of b_{1g}^2 and $b_{1g}e_u$ configurations, respectively. In other words, one implies two near equivalent locations for the additional hole, either to the Cu3d-O2p hybrid $b_{1g}(d_{x^2-y^2})$ state to form Zhang-Rice (ZR) singlet $^1A_{1g}$, or to purely oxygen nonbonding doublet $e_{ux,y}$ state with peculiar Cu^{2+}-Cu^{3+} valence resonance. The e_u hole can be coupled with the b_{1g} hole both antiferro- and ferromagnetically. This simple consideration indicates clearly a necessity to incorporate in the valence multiplet both the spin singlet $(b_{1g}e_u)^1E_u$ and the spin triplet $(b_{1g}e_u)^3E_u$, which energy could be even lower due to ferromagnetic $b_{1g}-e_u$ exchange. The pseudo-Jahn-Teller (JT) polaronic nature of the spin-singlet $^1A_{1g}-{}^1E_u$ ground state [14] favors their localization. In addition, one should account for the antiferromagnetic background which leads to the crucial enhancement of the effective mass for the moving spin singlets. So, a spin-singlet small pseudo-JT polaron as a hole ground state is likely to be immobile. In such a situation the most effective channel for the hole transport could be related to the low-lying excited spin-triplet $b_{1g}e_u : {}^3E_u$ term. This gives rise to a thermo-activated hole conductivity actually observed in most of slightly doped cuprates. The doped electron in cuprates occupies the only $b_{1g}(d_{x^2-y^2})$ orbital to form ZR-singlet-like $^1A_{1g}$ state. At first sight it implies the electron-hole symmetry of transport properties in cuprates, however, the existence of alternative e_u state for the hole actually points to strong electron-hole asymmetry. It should be noted that the nature itself of effective charge carriers in cuprates may evolve with doping from a single hole (electron) for lightly doped systems to a collective carrier like a charge density wave for a sizeable doping [15, 16].

In contrast to YBaCuO and LaCuO cuprates in the NdCeCuO and other electron doped systems we deal with an additional factor governing the quasiparticle transport. The matter concerns the rare-earth sublattice which is believed to strongly affect the low-temperature transport properties [6, 10]. Indeed, in frame of a simple activation mechanism of the conductivity, it is reasonable to assume that the activation is accompanied by a change in a crystal field acting on the R-ion and its magnetic anisotropy. Tetragonal crystal field includes the magnetic field-dependent part which can be written for the in-plane directed field as follows (see e.g., Ref.[17]):

$$H_{cf} = d(H_{\text{ext}})\cos 4\phi\,, \tag{1}$$

where ϕ is an azimuthal angle of the orientation of effective magnetic field for the Nd ion that is the sum of the external magnetic field and exchange fields due to Nd-Cu and Nd-Nd coupling. Thus we may conclude that the R-ion can indirectly affect the quasiparticle transport through activation energy which may intricately depend on the orientation of the surrounding Nd-ion magnetic momenta giving rise to a specific mechanism of anisotropic magnetotransport.

The explanation of low-temperature magnetotransport properties in $Nd_{2-x}Ce_xCuO_{4+\delta}$ system is hampered by the lack of information sufficient to understand and describe its magnetic state. To the best of our knowledge the magnetic-field studies were performed only for undoped Nd_2CuO_4, where the Cu sublattice first orders antiferromagnetically into the noncollinear type-I spin structure below $T_{N1}\approx 275\,\text{K}$ [18]. On further cooling, the Cu spins reorient into type-II (at $T_{N2}\approx 75\,\text{K}$) and type-III ($T_{N3}\approx 30\,\text{K}$) phases. In the type-II phase all the Cu spins rotate by $\pm 90^\circ$ about the c axis from the type-I phase. They rotate back to their original direction below T_{N3} in the type-III phase. As discussed in Ref.[17], we may consider the entire system to be built up of weakly interacting sets of planes, each set consisting of a Cu plane with one Nd plane above it and another below it. Thus, for most purposes it suffices to consider a 2D model consisting of a single set of Nd-Cu-Nd threeplanes. The energy of inter-plane A-B coupling may be written as a result of effective pseudo-dipolar interactions between Cu moments as follows:

$$E_{AB} = -Q\sin(\Phi_A+\Phi_B),$$

where $\Phi_{A,B}$ are the azimuthal angles of the orientation of Cu antiferromagnetic vectors for A, B units, respectively. This energy is minimal at $(\Phi_A+\Phi_B)=\pi/2$, if $Q > 0$, and at $(\Phi_A + \Phi_B) = -\pi/2, 3\pi/2$, if $Q < 0$. The parameter $Q > 0$ in phases I and III, and $Q < 0$ in phase II, in other words it changes the sign twice, that one explains as a result of a subtle competition of Cu-Cu, Cu-Nd, and Nd-Nd contributions to Q [17, 19]. It should be noted that any of these spin-reorientation transitions proceeds via two equivalent clockwise and counterclockwise rotations of Cu antiferromagnetic vectors when the relation $(\Phi_A + \Phi_B) = \pi/2$ (phases I,III) transforms into $(\Phi_A + \Phi_B) = -\pi/2, 3\pi/2$ (phase II).

On further lowering temperature, the large exchange coupling between the Cu and Nd polarizes the Nd spins and induces an ordered moment. Below about 30 K, the ordered arrangement of the Cu and Nd spins is noncollinear. At low temperatures $T < 1.5$ K, the application of a magnetic field of about 4.4 Tesla along the [100] direction seems to result in a spin-flop transition, in which the Cu and Nd spins oriented along the [100] direction rotate in-plane by 90° [20]. This would result in a collinear spin structure in which all of the ordered moments are approximately perpendicular to $\mathbf{H}_{\text{ext}}$. If a smaller field $(0.7 - 2\,\text{T})$ is applied along the [110] direction, there is a spin-flop transition in which all of the spins rotate in-plane by about 45°, giving rise to the same collinear spin structure [20]. The field, at which each of these transitions occurs, has been studied using several techniques, however the effect of an applied field with arbitrary direction in ab-plane remains to be unclear despite several theoretical attempts. The spin configuration of the noncollinear spin structure in NdCuO system has been analyzed in magnetic field parallel to the (ab) plane by Petitgrand et al. [19]. For the field along the [110] direction the transition is of second order with a critical spin-flop field $H_{SF}^{[110]}$. For $H < H_{SF}^{[110]}$, Cu-AFM vectors in the A and B units are expected to be oriented symmetrically with respect to the direction of external field at the angle [19, 13]

$$\Phi_A = \pi/2 - \Phi_B = -\frac{1}{2}\arcsin\frac{H^2 \sin 2\phi}{(H_{SF}^{[110]})^2}. \qquad (2)$$

However, it is worth noting that the angular dependence (2) seems to be applicable only in a rather narrow range near [110] direction. For the external field along the [100] direction the transition is of first order with a critical spin-flop field $H_{SF}^{[100]} > H_{SF}^{[110]}$. With increasing the angle inbetween the [100] direction and the external field the spin flop remains the first-order transition and smoothly approaches the second order when the external field approaches the [110] direction. However, the first-order transition for the external field turned slightly away from the [100] direction is accompanied by a relevant rotation of Cu-AFM vectors in the A and B units [19]:

$$\Phi_A = \pi/2 - \Phi_B \approx -\frac{H^2}{(H_{SF}^{[110]})^2}\phi. \qquad (3)$$

The magnetic data on Ce-doped system is very scant. Neutron-diffraction data [21] point to $T \approx 1.2$ K as the Nd-ordering temperature in single-crystalline sample $Nd_{2-x}Ce_xCuO_{4+\delta}$ $(x = 0.15)$ that is close to $T_{N_2} \approx 1.2$ K in Nd_2CuO_4. The dilution of Nd sublattice is expected to result in a considerable shift of spin-reorientation transitions I-II and II-III to low temperatures.

Recent neutron studies on Pr_2CuO_4 single crystals [22] have revealed rather unexpected spin structure transformations in differently in-plane directed external magnetic field that may result in a revision of many earlier magnetic data for $Nd_{2-x}Ce_xCuO_{4+\delta}$ as well. First of all, the authors emphasized the quantum character of spin system and orientational phase transitions. They have observed a novel phase transition from noncollinear phase with orthogonal AF subsystems to collinear one with spins oriented along [110] direction and proposed the transition seems to look like a conventional second order spin-flop transition only for the external magnetic field $\mathbf{H} \parallel [110]$, while with deviation from [110] direction the critical field rises, and the transition becomes the first order one for $\mathbf{H} \parallel [100]$ with critical field $H_C[100] = 5.4$ T. In all the cases, except $\mathbf{H} \parallel [110]$, the spin-flop state is reached only at $H \to \infty$.

Thus, the spin-flop transition in $Nd_{2-x}Ce_xCuO_{4+\delta}$ for the external field rotated in ab-plane is remarkable for a first-to-second order crossover which can manifest itself in AMR through a competition of two characteristic angular patterns typical for two orientations of external field, near two high symmetry directions, [100] and [110], respectively. In the absence of appreciable hole contribution for electron doped $Nd_{2-x}Ce_xCuO_{4+\delta}$ we may speculate that the main mechanism of magnetoresistance anisotropy is specified by the field dependence of activation energy for the electron transport due to the effect of field-dependent Nd-ion crystal field (1). But for all that we should take into account the nonequivalence of Nd ions from different sublattices with complex relation between the magnitude and orientation of effective fields on Nd ions and that for external magnetic field. Indeed, for small external magnetic field the Nd ions feel mainly the magnetic polarization of Nd-Cu system somehow or other ordered by external field, while for large magnetic field the Nd ions feel mainly the magnitude and orientation of external field. In such a case, the magnitude of exchange field on Nd ion appears as a peculiar border between low-field and high-field magnetotransport behavior. We suppose the unusual transformation of angular AMR pattern seen in Fig.3 and *reentrant* behavior of magnetoresistance at $T = 1.4\,\mathrm{K}$ seen in Fig.4 mirror a first-second order spin-flop crossover effect from the one hand, and a specific rare-earth contribution to the magnetotransport from the other hand. The external field $H_{\mathrm{ext}} \approx 4.0\,\mathrm{T}$ ($T = 1.4\,\mathrm{K}$) which defines a fairly narrow range of fields with a distinctly visible splitting effect and a remarkable suppression of magnetoresistance anisotropy is believed to provide a reliable estimate of exchange field on Nd ions. Interestingly, the 4.2 K data (Fig.2) reveal a fairly visible lobe's broadening with increasing the magnetic field, which may be addressed to be also a manifestation of remarkable nonequivalence of Nd ions from different sublattices for the external field rotated in ab-plane.

In conclusion, the nominally electron-doped $Nd_{2-x}Ce_xCuO_{4+\delta}$ ($x = 0.12$) is shown to reveal a strong low-temperature anisotropy of in-plane magnetoresistance. Specific fourfold angular magnetoresistance anisotropy reaching the magnitude of several percents was observed in oxygen annealed $Nd_{2-x}Ce_xCuO_{4+\delta}$ films at low temperatures and in an external magnetic field up to 5.5 T. At low temperature $T = 1.4\,\mathrm{K}$ we observed the unusual transformation of magnetoresistance response with increasing the external magnetic field which seems to be a manifestation of a combined effect of a crossover between first and second order spin-flop transitions and a field-dependent rare-earth contribution to quasiparticle magnetotransport. At present, we have uncovered only a part of features which characterize the AMR in NCCO system. Unfortunately, the lack of the detailed magnetic and transport information does not allow a full quantitative description of the effect. The unified and unambiguous interpretation of all the aspects of this complex effect remains a challenging problem for a future experimental research and theoretical studying.

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