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Giant anisotropy of the magnetoresistance and the 'spin valve' effect in antiferromagnetic Nd$_{2-x}$Ce$_x$CuO$_4$

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Abstract

We have studied anisotropic magnetoresistance (MR) and magnetization with a rotating magnetic field (B) within the CuO$_2$ plane in lightly doped AF Nd$_{2-x}$Ce$_x$CuO$_4$. *A giant anisotropy* in the MR is observed at low temperature, below 5 K. The *c*-axis resistivity can be tuned over about one order of magnitude just by changing the B direction within the CuO$_2$ plane, and a scaling behavior for the out-of-plane and in-plane MR is found. A 'spin valve' effect is proposed for explaining the giant anisotropy of the out-of-plane MR and the evolution of the scaling parameters with the external field. It is found that the field-induced spin-flop transition of the Nd^{3+} layer under high magnetic field is the key to understanding the giant anisotropy. These results suggest that a novel entanglement of charge and spin dominates the underlying physics.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

It is generally believed that the pairing necessary for high-T_c superconductivity in cuprates involves interplay between doped charges and antiferromagnetic (AF) spin correlation. In this sense, the study of the lightly doped, insulating AF state is important to understanding the pairing mechanism because the density of the carriers can be sufficiently low that the interaction between them is small relative to their interaction with Cu^{2+} spins. Many intriguing and anomalous phenomena have been observed for lightly doped AF cuprates due to strong coupling between charges and Cu^{2+} spins [1–5]. Cu^{2+} spins order in an AF collinear structure for the parent compounds of hole-doped cuprates [6, 7], while they order in an AF noncollinear structure for those of electron-doped cuprates [8, 9]. All spins point either parallel or antiparallel to a single direction in AF collinear structure, while the spins in

adjacent layers are orthogonal in AF noncollinear structure. A transition from a noncollinear to a collinear spin arrangement with a spin flop can be induced by a certain magnetic field (B_c) [10], which is confirmed for lightly electron-doped Pr$_{1.3-x}$La$_{0.7}$Ce$_x$CuO$_4$ [4] and Nd$_{2-x}$Ce$_x$CuO$_4$ [5] crystals, and such a transition significantly affects both the in-plane and the out-of-plane resistivity.

In Nd$_2$CuO$_4$, the Cu^{2+} spins order in three phases with two different AF noncollinear spin structures and experience two reorientation phase transitions [8, 11–13]. It has been reported by us that MR anisotropy with a fourfold symmetry in different AF spin structures upon rotating the magnetic field (B) within the *ab*-plane is observed in lightly doped Nd$_{2-x}$Ce$_x$CuO$_4$ above 10 K [5], while MR anisotropy with a twofold symmetry is seen at the spin reorientation temperatures. A large anisotropic MR was observed in lightly electron-doped Pr$_{1.3-x}$La$_{0.7}$Ce$_x$CuO$_4$ [4] and Nd$_{2-x}$Ce$_x$CuO$_4$ [5]. These results indicate strong spin–charge coupling in electron-doped

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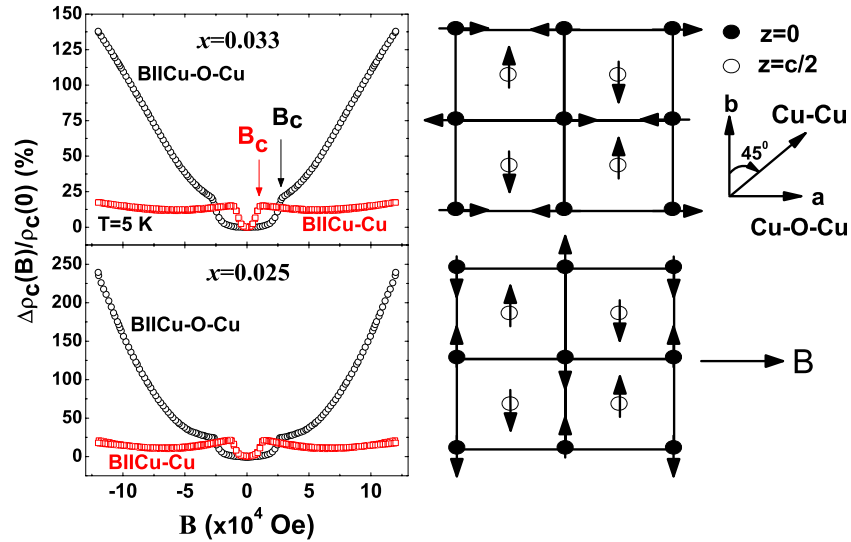[3] Author to whom any correspondence should be addressed.

Figure 1. Isothermal MR at 5 K with B along the Cu–O–Cu and Cu–Cu directions for the samples $Nd_{2-x}Ce_xCuO_4$ with $x = 0.025$ and 0.033, respectively. Zero-field noncollinear spin structure; only Cu spins are shown; field-induced transition from noncollinear to collinear spin ordering with B along the Cu–O–Cu direction.

cuprates. In Nd_2CuO_4, the magnetic coupling between Nd^{3+} and Cu^{2+} is very important at low temperature since the magnetic moment of Nd^{3+} becomes large with decreasing temperature ($\sim 1.3 \mu_B$ at 0.4 K) [12]. Magnetic structures of Nd^{3+} are very abundant at low temperature [14–16]. In this sense, electronic transport at low temperature is expected to be sensitive to change of the magnetic structure of Nd^{3+} due to strong spin–charge coupling. This will provide us with a chance to understand the spin–charge coupling in electron-doped cuprates. The lightly electron-doped cuprates represent a good system in which to study the coupling between charge and Cu^{2+} spin because: (1) the spin structure can be tuned by an external magnetic field [10]; (2) in contrast to the buckled CuO_2 in hole-doped cuprates, the CuO_2 plane in electron-doped cuprates is flat, so the spin ordering is pure antiferromagnetic without a ferromagnetic component along the c-axis in hole-doped cuprates; such a ferromagnetic component along the c-axis makes the study of the coupling between charge and Cu^{2+} spin complicated. In this work, we study angle dependent magnetoresistance and magnetization below 10 K in lightly electron-doped $Nd_{2-x}Ce_xCuO_4$. *A giant anisotropy* in MR is observed, and the c-axis resistivity can be tuned over about one order of magnitude just by changing the B direction. The scaling behavior for in-plane and out-of-plane MR is systematically changed with increasing magnetic field. The jump in the MR with B around the Cu–O–Cu direction coincides with the sudden change in magnetization at low temperature below 5 K. The underlying physics will be discussed below.

2. Experiment

Growth of single crystals and their resistivity have been reported in previous work [5]. The susceptibility and magnetoresistance were measured with a superconducting quantum interference device (SQUID) with a 7 T maximal

magnetic field and a Quantum Design PPMS system with a 12 T maximal magnetic field, respectively. In our measurements, the maximal magnetic field is 12 T for MR and 7 T for magnetization. ρ_{ab} and ρ_c stand for in-plane resistivity and out-of-plane resistivity, respectively. The magnetoresistance is defined as MR $= \frac{\Delta \rho(B)}{\rho(0)} = \frac{\rho(B)-\rho(0)}{\rho(0)}$. It should be noted that all results discussed below are well reproducible.

3. Results and discussion

Figure 1 shows the isothermal out-of-plane MR at 5 K for single crystals with $x = 0.025$ and 0.033 with B along the Cu–Cu and Cu–O–Cu directions, respectively. The MR behavior is similar to that observed for antiferromagnetic $Pr_{1.3-x}La_{0.7}Ce_xCuO_4$ with an $x = 0.01$ crystal [4]. But the magnitude of the MR and the MR anisotropy are much larger than in the case of $Pr_{1.3-x}La_{0.7}Ce_xCuO_4$. The step-like increase of MR corresponds to the noncollinear–collinear transition occurring at the critical field B_c. As shown in figure 1, the critical field B_c along the Cu–O–Cu direction is larger than that along the Cu–Cu direction. Above B_c, the behavior of MR for B along the Cu–Cu direction is quite different from that for B along the Cu–O–Cu direction in the collinear structure. The MR with B along the Cu–Cu direction slightly changes above B_c, while the MR monotonically increases with increasing B for B along the Cu–O–Cu direction. A giant anisotropic MR between the fields B along the Cu–Cu and Cu–O–Cu directions is observed. For the $x = 0.025$ crystal, the MR at 12 T is as high as $\sim 235\%$ with B along the Cu–O–Cu direction, while it is only $\sim 17\%$ with B along the Cu–Cu direction.

Upon rotating a magnetic field larger than B_c within the CuO_2 plane, the spins always keep the collinear arrangement and the spin structure rotates as a whole; all spins are perpendicular to the magnetic field as shown in figure 1 [10].
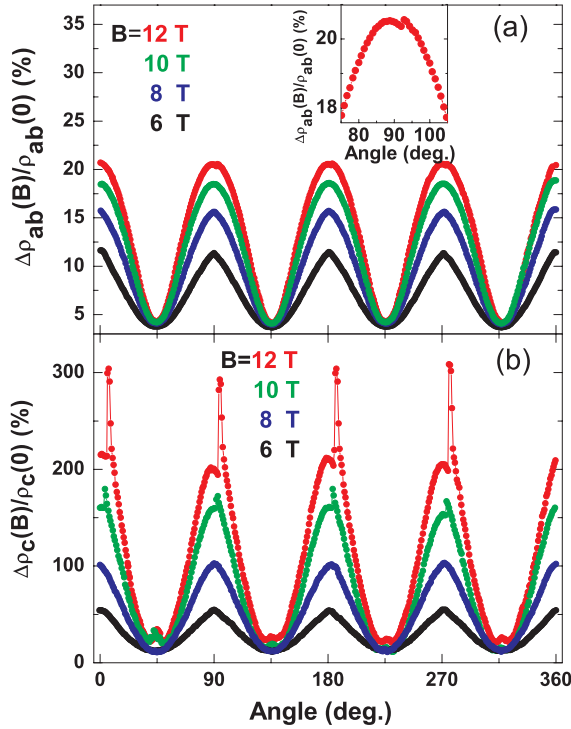
Figure 2. (a) Isothermal in-plane and (b) out-of-plane MR at 5 K under different B fields as a function of the angle between B and the Cu–O–Cu direction upon rotating B within the CuO$_2$ plane for the single crystal with $x = 0.025$. The inset in (a): magnified in-plane MR with $B = 12$ T.

Figure 3. Out-of-plane MR as a function of the angle between B and the Cu–O–Cu direction upon rotating B within the CuO$_2$ plane at 2 K, 4 K and 5 K, for the single crystal with $x = 0.033$ ($B = 12$ T).

In order to study the anomalous and giant anisotropic MR, we carefully investigated the evolution of in-plane and out-of-plane MR with rotating B within the CuO$_2$ plane. Figures 2(a) and (b) show the evolution of the in-plane and out-of-plane MR with the angle between B and the Cu–O–Cu ([100]) direction at 5 K for the single crystal with $x = 0.025$. Both the in-plane and the out-of-plane MR increase with increasing B, and show a giant anisotropy with fourfold symmetry; such fourfold symmetry arises from the symmetry of magnetic structure because there exist two equivalent spin easy axes (Cu–Cu direction) and two equivalent spin hard axes (Cu–O–Cu direction) in the collinear spin structure, which has been confirmed by the different critical fields B_c for B along the Cu–O–Cu and Cu–Cu directions as shown in figure 1. A striking feature is observed: that the out-of-plane MR at 12 T sharply increases from ∼200% to ∼300% at an angle close to B along Cu–O–Cu. Such behavior originates from the spin flop induced by the magnetic field with B close to the Cu–O–Cu direction as discussed below. A similar jump can also be observed at an angle close to B along Cu–Cu, but the jump is very small compared to the case for B close to the Cu–O–Cu direction.

In order to study the effect of temperature on the anisotropy of MR, we systematically investigated the MR behavior of the $x = 0.033$ crystal because the resistivity of the $x = 0.025$ crystal is too large to be measured due to the resistivity divergence at low temperature. Figure 3 shows evolution of the out-of-plane MR upon rotating B within the CuO$_2$ plane at 2, 4 and 5 K under 12 T for the $x = 0.033$
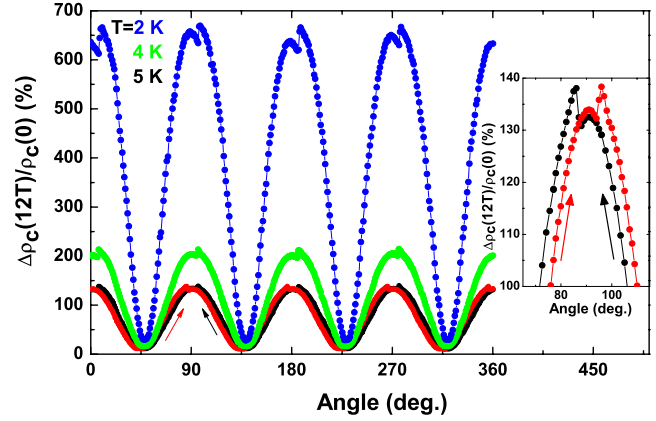
crystal. The results are similar to that observed for the crystal with $x = 0.025$. The MR increases monotonically and the anisotropy of the MR induced by rotating B within the Cu–O plane apparently increases with decreasing temperature. The MR under 12 T with B along the Cu–Cu direction is about 11.2% at 5 K, 17.1% at 4 K and 27.7% at 2 K; while the MR with B along the Cu–O–Cu direction is about 133% at 5 K, 203% at 4 K and 656% at 2 K. This indicates that a giant anisotropy of the resistivity is induced by a magnetic field with B along Cu–O–Cu and Cu–Cu at low temperature. At 2 K, the resistivity under 12 T with B along the Cu–O–Cu direction is about one order of magnitude larger than that with B along the Cu–Cu direction. Such giant anisotropy in resistivity induced just by changing the B direction within the CuO$_2$ plane should be related to the magnetic structure and magnetic moment induced by B, because the magnetic field along Cu–O–Cu or Cu–Cu just changes the spin structure and induces different magnitudes of the magnetic moment. To understand the jump in the MR with B close to the Cu–O–Cu direction, the MR at 5 K is measured while rotating B in the clockwise direction and in the anticlockwise direction. It is found that the MR jumps observed with B rotating in the clockwise direction and in the anticlockwise direction are symmetric relative to B along the Cu–O–Cu direction, as shown in the inset of figure 3. This indicates that the spin does not prefer the Cu–O–Cu direction, and the spin jump always occurs around the Cu–O–Cu direction when B is rotated within the CuO$_2$ plane. Therefore, the jump arises from the spin flop induced by B.

In figure 4, the same data for in-plane and out-of-plane MR as were shown in figures 2(a) and (b) are plotted as $\Delta\rho_c(B)/\rho_c(0)$ as a function of $\Delta\rho_{ab}(B)/\rho_{ab}(0)$. Only the data for in-plane and out-of-plane MR between 45° and 90° are plotted in figure 4 because the in-plane and out-of-plane MR exhibit exactly the same oscillation. All data above can be fitted with $\mathrm{MR}_c = \beta + \alpha \cdot \mathrm{MR}_{ab}^v$ very well. The fitting parameters are listed in table 1. It is found that the fitting parameter β is zero below 10 T. The fitting parameter v increases from ∼1 to ∼3 with increasing magnetic field. These results indicate that the relation between out-of-plane MR and in-plane MR is strongly dependent on the external
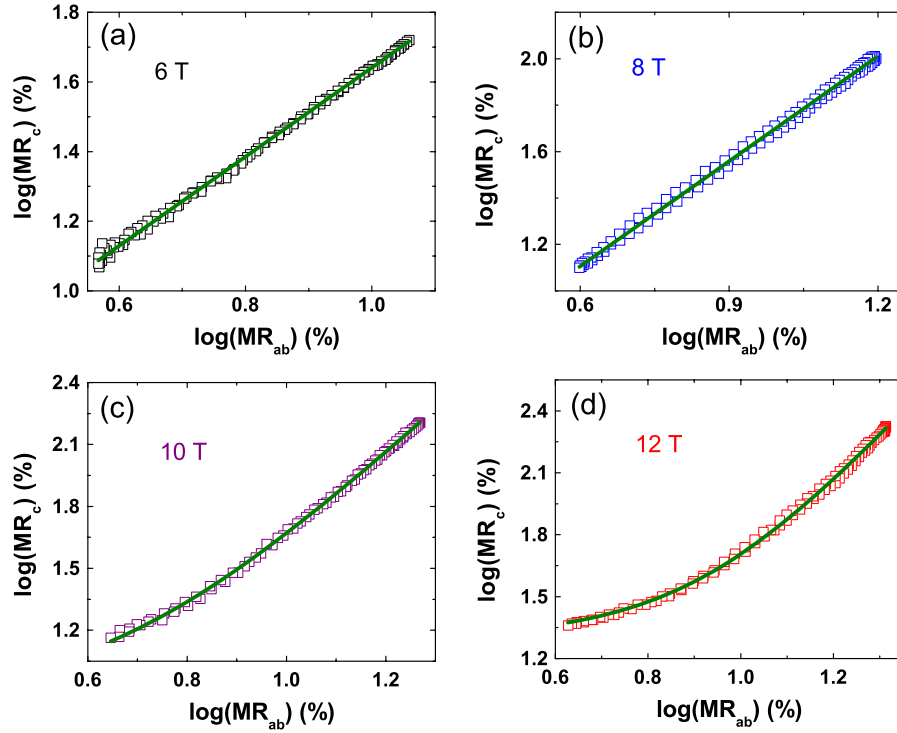
Figure 4. The same data as were shown in figure 2 are plotted as $\Delta\rho_c(B)/(\rho_c(0))$ as a function of $\Delta\rho_{ab}(B)/(\rho_{ab}(0))$. The line is the fitting result, with the formula $\Delta\rho_c(B)/\rho_c(0) = \beta + \alpha(\Delta\rho_{ab}(B)/\rho_{ab}(0))^\upsilon$. At 6 and 8 T, the parameter $\beta = 0$.

Table 1. Fitting parameters α, β and υ with the formula $\Delta\rho_c(B)/\rho_c(0) = \beta + \alpha(\Delta\rho_{ab}(B)/\rho_{ab}(0))^\upsilon$ under different fields.

Field (T)	β	α	υ
6	0	2.30	1.29
8	0	1.59	1.51
10	7.47	0.25	2.20
12	20.13	0.10	2.50

magnetic field. It is suggested that the out-of-plane and in-plane transport are closely related to the magnetic structure since the external magnetic field can modify the spin structure. The giant anisotropy could arise from the change of spin structure induced by the external magnetic field.

In order to further understand how magnetic field influences the transport, understanding the evolution of magnetic structure under a magnetic field is very necessary. Figure 5 shows the magnetization under 7 T at 2 K with B rotating within the CuO_2 plane for the AF $Nd_{2-x}Ce_xCuO_2$ with $x = 0$, 0.025, 0.06 and 0.13. It is found that the magnetization shows the same fourfold symmetry with B rotating within the CuO_2 plane as is observed for the MR shown in figure 2. The amplitude of the oscillation and the magnetization decrease with increasing x. A striking feature is observed: that the magnetization shows a jump with B around the Cu–O–Cu direction at which a corresponding jump is observed in the MR, as shown in the inset of figure 5. However, this fourfold symmetry gradually disappears with increasing temperature, as shown in figure 6. As we know, the magnetic moment of Nd^{3+} increases prominently at low temperature, and the magnetization is very sensitive to the
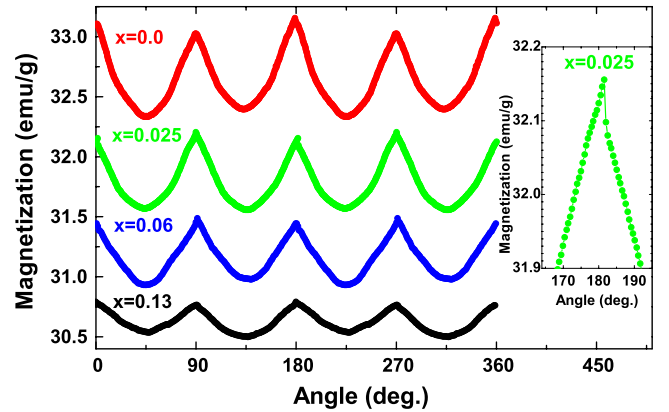

Figure 5. The magnetization measured under a magnetic field of 7 T at 2 K as a function of the angle between B and the Cu–O–Cu direction upon rotating B within the CuO_2 plane for the samples with $x = 0$, 0.025, 0.06, 0.13. The inset shows a jump in magnetization at a certain angle corresponding to the jump observed in the MR.

magnetic moment of Nd^{3+} below 5 K [14, 17]. Therefore, the fourfold symmetry in magnetization and the jump in magnetization are related to the magnetic structure of Nd^{3+}. On the other hand, the fourfold symmetry shown in figure 5 indicates a magnetic ordering of Nd^{3+}. The spontaneous ordering of the Nd^{3+} subsystem at low temperature due to Nd^{3+}–Nd^{3+} interaction remains controversial. X-ray magnetic scattering data indicate that Nd^{3+} ions are polarized at 37 K [18]. The removal of the Kramers doublet degeneracies observed using crystal field infrared transmission indicates
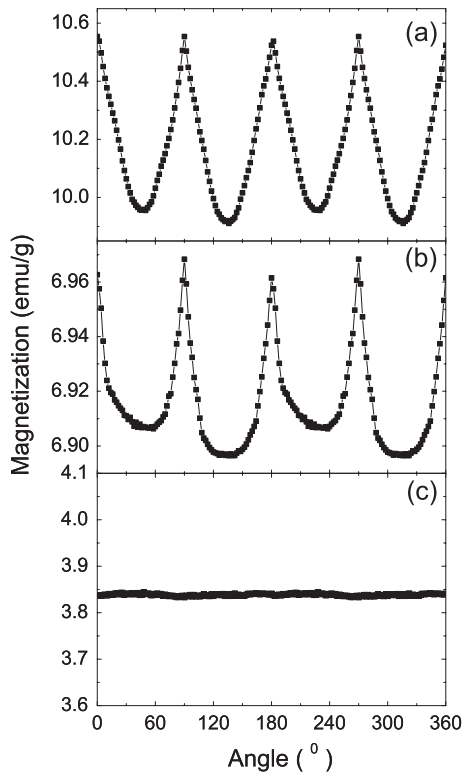
Figure 6. The increment of magnetization measured under a magnetic field of 6.5 T at (a) 2 K, (b) 3.5 K and (c) 5 K relative to the magnetization at 7.5 K as a function of the angle between *B* and the Cu–O–Cu direction upon rotating *B* within the CuO_2 plane for the samples with $x = 0.025$.

that these ions are already polarized by the Cu^{2+} subsystem at a temperature as high as 140 K [19]. An enhancement of the neutron scattering magnetic peak intensities around 3 K has been interpreted as attributable to Nd^{3+} ordering due to Nd^{3+}–Nd^{3+} interaction [12], while Lynn *et al* have estimated the Nd^{3+} ordering temperature around 1.5 K [17]. Recently, an abnormal peak around 5 K observed in ultrasonic measurements was explained as being somehow related to local magnetic domains [15]. Since the Nd^{3+}–Cu^{2+} and Nd^{3+}–Nd^{3+} interactions are opposite, the former is dominant above 5 K and makes Nd^{3+} parallel to Cu^{2+}, as shown

in figure 8(a), while the later is dominant below 5 K and makes Nd^{3+} 'prefer' to be perpendicular to Cu^{2+}, as shown in figure 8(b). Due to the frustration of the Nd^{3+} magnetic subsystem arising from the competition between Nd^{3+}–Cu^{2+} and Nd^{3+}–Nd^{3+} interactions, the local magnetic domain is formed with Nd^{3+} not parallel to the Cu^{2+} magnetic moment below 5 K. The magnetic structure of the Nd^{3+} subsystem with a magnetic moment of Nd^{3+} perpendicular to Cu^{2+} can be stabilized by the external field. The observed fourfold symmetry below 5 K in the magnetization could arise from the reorientation of the Nd^{3+} spin. Richard *et al* have provided evidence that the magnetic structure below 5 K has anisotropic field dependence [15]. As shown in figure 7, the field dependent magnetization of the $x = 0.025$ sample at 2 K shows an anomaly at 0.6 T and 3.6 T for Cu–Cu and Cu–O–Cu directions, respectively. However, no such anisotropy is observed above 5 K. To make the anisotropy clear, the magnetization at 2 K subtracted from the 5 K magnetization is shown in figure 7(b). Such an anomaly has been attributed to spin reorientation of Nd^{3+} in Nd_2CuO_4 [14]. The spin reorientation of Nd^{3+} occurs due to a transition from the magnetic structure shown in figure 8(a) to that shown in figure 8(b). It is surprising that the magnetization for two directions has a crossing around 6 T and the magnetization is somewhat saturated, as shown in figure 7. It is suggested that the magnetic structure under high magnetic field is different from that under low magnetic field. A similar result has been reported for Nd_2CuO_4 [14]. Recently, a crossover from an antiferromagnetic to a paramagnetic configuration induced by high magnetic field was proposed by Richard *et al* [16]. The corresponding magnetic structures given by Richard *et al* are shown in figures 8(c)–(f). When the in-plane magnetic field $B < 4$ T is along the Cu–O–Cu and Cu–Cu directions, the collinear magnetic structures are as shown in figures 8(c) and (e), respectively. In the configuration, the Cu^{2+}–Nd^{3+} interaction is larger than the Nd^{3+}–Nd^{3+} interaction. When the in-plane magnetic field $B > 4$ T is along the Cu–O–Cu and Cu–Cu directions, the Nd^{3+}–Nd^{3+} interaction is dominant and the magnetic structure changes from an antiferromagnetic to a paramagnetic configuration as shown in figures 8(d) and (f), in which the Nd^{3+} spins are aligned in the applied magnetic field and thus behave as ferromagnetic-like. The crossing at about 6 T in magnetization could be related to the change of magnetic structure shown in figure 8.
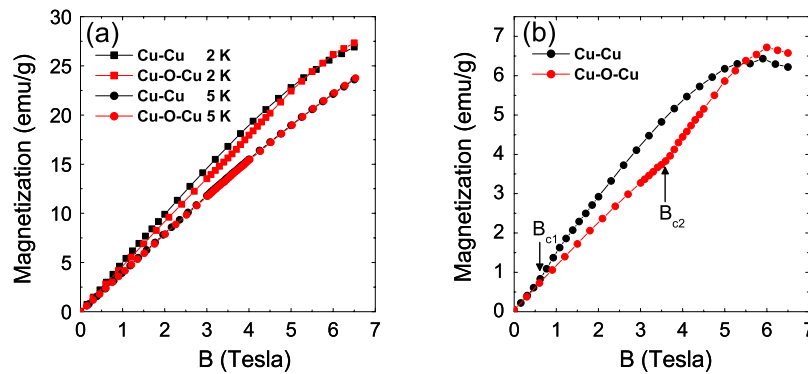

Figure 7. (a) Magnetization as a function of magnetic field along the Cu–Cu and Cu–O–Cu directions at 2 K and 5 K; (b) $M(2\,\text{K}) - M(5\,\text{K})$.
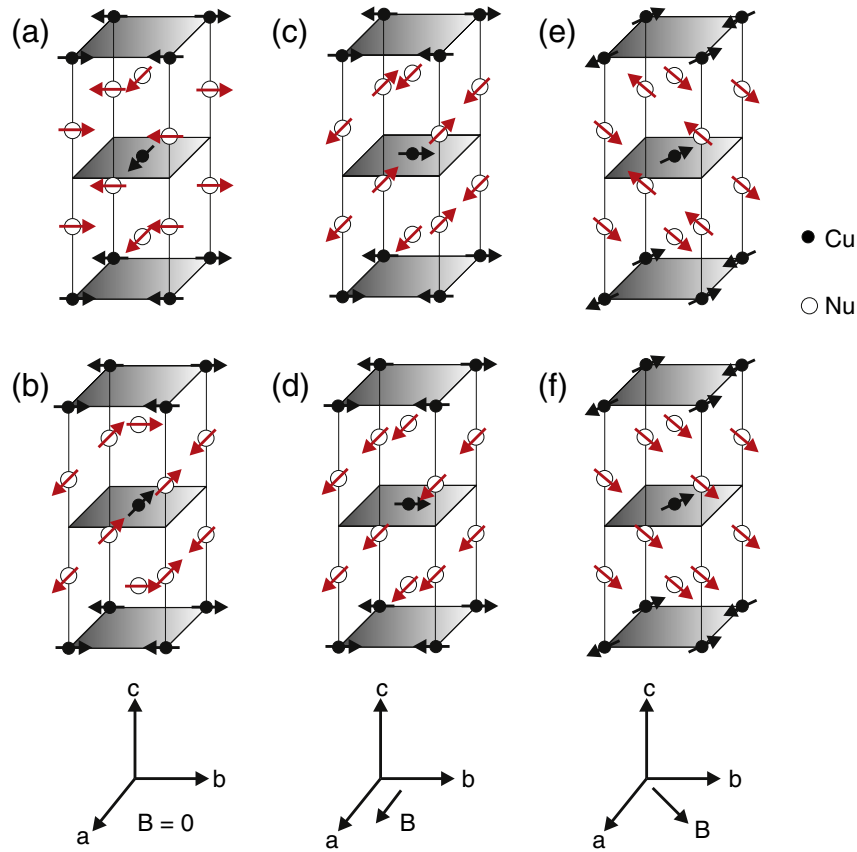
Figure 8. Magnetic configuration of Nd_2CuO_4 in the noncollinear antiferromagnetic phase: (a) above 5 K, (b) below 5 K; the magnetic configuration of Nd_2CuO_4 in the collinear phase assuming an antiferromagnetic alignment of Nd^{3+} spins: (c) $B \parallel [100]$, (e) $B \parallel [110]$; and a ferromagnetic-like alignment of the Nd^{3+} spins: (d) $B \parallel [100]$, (f) $B \parallel [110]$.

The fourfold symmetry in the magnetoresistance has been observed above 5 K in the same sample in previous work [5], while similar symmetry in magnetization arising from Nd^{3+} spins is observed only below 5 K. Therefore, the fourfold symmetry in the MR should arise from anisotropic magnetic structure of Cu^{2+}, as discussed in our previous work [5]. As shown in figures 2 and 6, the giant anisotropy in the MR below 5 K coincides with the magnetic ordering of Nd^{3+} with the same fourfold symmetry. These results indicate that the fourfold symmetry of the MR results from spin ordering of the Cu^{2+}, and the spin ordering of Nd^{3+} enhances the fourfold symmetry in the MR below 5 K and leads to a giant anisotropy in the MR. It is evident that the jump of out-of-plane MR with B along the Cu–O–Cu direction shown in figure 2 is related to the sudden change in magnetization shown in the inset of figure 5. Therefore, the change of MR below 5 K relative to the high temperature MR can be mainly ascribed to the ordering of Nd^{3+} spins. As shown in figure 4, there exists a scaling behavior for the out-of-plane MR and in-plane MR with $MR_c = \beta + \alpha MR_{ab}^v$. But the fitting parameters β, α and v show a systematic change with increasing external field, as listed in table 1. Change of the scaling behavior is closely related to the change of the magnetic structure of Nd^{3+} induced by the external field since the increase of the external field from 6 to 12 T cannot lead to change of the magnetic structure of the Cu^{2+} subsystem. This is supported

by the fact that the change of the in-plane MR with increasing magnetic field is much smaller than that of the out-of-plane MR, and the anisotropy of the out-of-plane MR is much larger than that of the in-plane MR as shown in figure 2. Richard *et al* [16] pointed out that the magnetic structure of the Nd^{3+} subsystem can change from an antiferromagnetic to a paramagnetic configuration at a certain critical magnetic field. It is possible that the evolution of the scaling parameters listed in table 1 is related to the change of the magnetic structure of Nd^{3+}. It is well known that the neighboring CuO_2 plane with the antiferromagnetic configuration is separated by the Nd–O layer [15]. As discussed above, the magnetic structure of the Nd^{3+} subsystem can be turned by the external field. Therefore, the out-of-plane transport can be switched by the magnetic field, assuming the Nd–O layer as a barrier. In this sense, this phenomenon can be well understood with a 'spin valve' effect. The magnetic excitations are different with B along the Cu–Cu and Cu–O–Cu directions because the magnetic structure is more frustrated around Cu–O–Cu [15]. In the 'spin valve' picture, the different magnetic excitations in the Nd^{3+} layer lead to different transport along the c-axis. Therefore, the spin-flop transition is the key to understanding the giant anisotropy of the out-of-plane MR. Thermal conductivity results indicate that the in-plane magnetic field can result in a closing of the anisotropic gap ($\sim$0.3 meV) which leads to additional heat conduction [20]. The critical magnetic field is about 4.5 T and

2.5 T in the Cu–O–Cu and the Cu–Cu direction, respectively. The closure of the anisotropic gap corresponds to a spin-flop transition for Cu^{2+} spins. This result indicates that the spin-flop transition can lead to a closure of the anisotropic gap. At low temperature, below 5 K, another gap related to the spin-flop transition of Nd^{3+} is closed under high magnetic field and this gap is anisotropic. The gap can be estimated using $B = \Delta/g\mu_B$ ($B \sim 8$ T); the gap is about 0.5 meV with magnetic field along the Cu–O–Cu direction. This spin flop should originate from the Nd^{3+} subsystem because magnons from Cu^{2+} have energy above 5 meV [21–23], and four optical Nd magnon branches lie in the range 0.2–0.8 meV [24, 25]. Study on the magnetic structure under high magnetic field at low temperature is lacking. This picture needs further experimental investigation to confirm it. These results give strong evidence that a nontrivial correlation between charge and the AF ordering background exists; the charge transport can be affected not only by Cu^{2+} spins but also by Nd^{3+} spins, especially below 5 K.

The huge changes in resistivities as induced by the in-plane magnetic field seem to be a highly nontrivial phenomenon. Note that the applied in-plane magnetic field should only affect the spins of the system via the Zeeman effect without directly influencing the orbital motion of charge carriers in the in-plane case, and presumably with only a weak orbital effect for the out-of-plane case, as the resistivity itself is divergent at low temperature. This thus implies the existence of some kind of strong 'entanglement' of the spin and charge degrees of freedom such that tuning the magnetic ordering with an in-plane magnetic field can result in the big enhancement of resistivities seen in the measurements. Furthermore, the large MR behavior in this insulating regime also strongly suggests that the divergence of resistivities at low temperature may not be simply a conventional localization effect due to disorders since spin structures can affect resistivities so much. Although the microscopic mechanism remains unclear, the novel spin–charge entanglement does exist in strongly correlated models. For example, in the t–J model, a so-called phase string effect has been shown [26] to be present as a non-local mutual frustration of the charge and spin degrees of freedom induced by doped charge carriers moving in an antiferromanget. In fact, the localization of the charge carriers in the magnetic ordered phase has been interpreted [27] on the basis of such a phase string effect and it is thus conceivable that the change of the spin structure may strongly affect the resistivities via the phase string effect. The scaling for in-plane and out-of-plane MR is strongly dependent on the spin structure of Nd^{3+} which emerges at low temperature, since strong Cu^{2+}–Nd^{3+} interaction can be easily tuned by an external magnetic field. This provides a good chance to show evidence for the spin–charge entanglement.

4. Conclusion

In this paper, we study anisotropic magnetoresistance (MR) and magnetization with a rotating magnetic field (B) within the CuO_2 plane in lightly doped AF $Nd_{2-x}Ce_xCuO_4$. *A giant anisotropy* in the MR is observed, and the c-axis resistivity can

be tuned over about one order of magnitude just by changing the B direction. These results provide evidence to support the spin-flop transition of Nd^{3+} ions induced by a high magnetic field. The change of magnetic structure induced by different external fields leads to a systematic evolution of the scaling behavior for in-plane and out-of-plane MR. A 'spin valve' effect is used to explain clearly the out-of-plane MR behavior. Such novel entanglement of charge and spin dominates the underlying physics.

Acknowledgments

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