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# Unusual anisotropic magnetoresistance in charge-orbital ordered $Nd_{0.5}Sr_{0.5}MnO_3$ polycrystals

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Due to its potential application in magnetic recording and sensing technologies, the anisotropic magnetoresistance (AMR) effect has attracted lasting attention. Despite the long history, AMR effect has not been fully understood especially in the unconventional materials, such as perovskite manganites. Here, we report an unusual AMR effect in the charge-orbital ordered (COO) $Nd_{0.5}Sr_{0.5}MnO_3$ polycrystals, which is observed when the magnetic field rotates in the plane that is perpendicular to the current (out-of-plane AMR). Despite being a polycrystalline sample where no anisotropy is expected, the resistivity shows a large irreversible drop with rotating magnetic field. A model has been proposed based on anisotropic magnetic field induced the melting of COO phase to explain the unusual out-of-plane AMR successfully. Our results demonstrate a new way for understanding the close relationship between phase separation and AMR effect in COO manganites. © *2014 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4904437]

## I. INTRODUCTION

Anisotropic magnetoresistance (AMR) refers to the dependence of resistivity change on the direction of magnetic field applied. Despite its long history and successful application in magnetic recording technologies,[1,2] the AMR effect has not been fully understood especially in the unconventional ferromagnets, which has led to a renewed interest in this effect.[3–12] The AMR effect is usually measured by rotating the magnetic field ($H$) in the plane that contains both magnetic moment ($M$) and current ($I$) [in-plane AMR, Fig. 1(a)]. The resistivity ($\rho$) depends on the relative angle ($\alpha$) between $M$ and $I$ obeying $\rho = \rho_\perp + (\rho_\parallel - \rho_\perp)\cdot\cos^2\alpha$, where $\rho_\parallel$ and $\rho_\perp$ correspond to the resistivity with $M\|I$ and $M\perp I$, respectively. From a phenomenological point of view, AMR effect has a non-crystalline component where the resistivity depends on the relative angle between $M$ and $I$, and a crystalline component where the resistivity depends on the absolute orientation of $M$ and $I$ with respect to the crystal axes.[13,14] In 3$d$ ferromagnetic metals, the crystalline component is usually smaller than the non-crystalline component.[15] The in-plane AMR effect is usually interpreted by the spin-orbit coupling, which gives rise to a magnetization dependent $s$-$d$ scattering.[14,16,17]

In recent years, special attention has been paid to perovskite manganites where very large AMR has been observed.[5,18–23] The AMR shows different temperature and magnetic field dependence as compared to that in 3$d$ ferromagnetic alloys, suggesting different mechanisms behind. In contrast to that in 3$d$ ferromagnetic alloys, the crystalline component of AMR in perovskite manganites can be very large, and a large resistance change can be observed even by rotating $H$ in the plane that is perpendicular to $I$ [out-of-plane AMR, Fig. 1(b)].[19,20] Normally, for a polycrystalline sample where the crystalline anisotropy is averaged out, the out-of-plane AMR is expected to be zero. In this report, the AMR effect in COO $Nd_{0.5}Sr_{0.5}MnO_3$ polycrystals has been measured by utilizing the out-of-plane configuration. Despite being a polycrystal where no anisotropy is expected, notable irreversible resistance drop upon rotating $H$ in the COO state was observed, indicating an unusual AMR phenomenon that correlates with phase competition between COO and the field-induced FMM phase.[21–23] Due to the fact that there is strong crystalline orientation dependent magneto-transportation behavior in perovskite manganites,[19,20] a model based on anisotropic magnetic field induced first-order phase transition from the COO state to the ferromagnetic metallic (FMM) state has been proposed to explain the unusual AMR effect. The model can fit our results very well, thus demonstrating a possible way for understanding the close relationship between phase separation and AMR in charge-orbital ordered manganites.

## II. EXPERIMENTAL SECTION

$Nd_{0.5}Sr_{0.5}MnO_3$ polycrystals were prepared by the standard solid state reaction method. Stoichiometric mixture of high-purity $Nd_2O_3$, $SrCO_3$, and $MnO_2$ was grounded in an $Al_2O_3$ crucible and fired at 1000 °C for 24 h. The resulting powder was then sintered at 1250 °C and 1400 °C for 24 h with intermediate grindings, the powders was pressed into pellets under a pressure of 20 MPa before the final firing. The prepared $Nd_{0.5}Sr_{0.5}MnO_3$ ceramics were examined by X-ray diffraction and confirmed to be of single phase. Samples for electrical measurements were cut into both cuboid- and cylinder-shaped samples. Temperature dependent magnetization was measured

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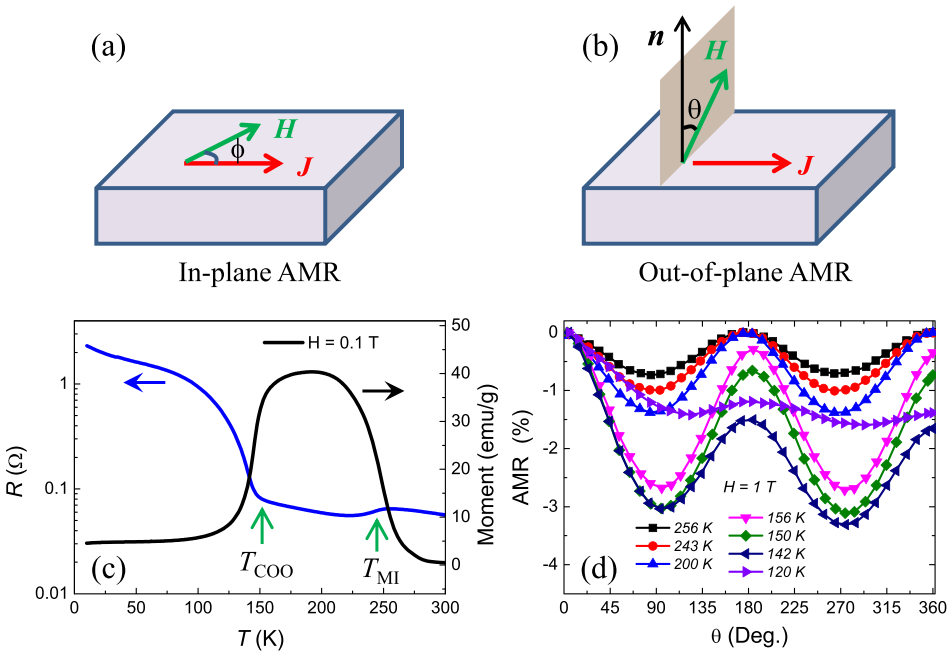


FIG. 1. (a) and (b) Schematic pictures showing two configurations used for measuring the AMR. (c) Temperature dependent resistance and magnetic moment of $Nd_{0.5}Sr_{0.5}MnO_3$ polycrystals, the metal-insulator transition temperature ($T_{MI}$) and charge-orbital ordering temperature ($T_{COO}$) are indicated by the arrows. (d) Anisotropic magnetoresistance measured at various temperatures, the magnetic field is fixed at 1 T.

using a Magnetic Property Measurement System (MPMS, Quantum Design). Magneto-transportation measurements were conducted using a Physical Property Measurement System (PPMS, Quantum Design) equipped with a motorized sample rotator, and the standard four-wire method was used for the resistance measurements. The out-of-plane AMR in COO $Nd_{0.5}Sr_{0.5}MnO_3$ polycrystals was measured and calculated following: $AMR = [R(\theta = 180°) - R(\theta = 0°)]/R(\theta = 0°)$. The AMR hereafter denotes the out-of-plane AMR, if not otherwise specified.

## III. RESULTS AND DISCUSSION

Figure 1(c) shows the resistance and magnetization of the $Nd_{0.5}Sr_{0.5}MnO_3$ polycrystals as a function of temperature. Consistent with earlier reports,[24] there is a metal-insulator transition (MIT) from paramagnetic insulating (PI) state to ferromagnetic metallic (FMM) state at $T_{MI} \sim 243$ K, and the antiferromagnetic insulating (AFI) COO state shows up below $T_{COO} \sim 156$ K. The FMM phase is found to persist below 156 K, coexisting with the COO phase in the low temperature region. Figure 1(d) shows the AMR in $Nd_{0.5}Sr_{0.5}MnO_3$ polycrystals at different temperatures. Interestingly, the AMR shows different angular dependence as temperature is varied. For $T > T_{COO}$, the AMR follows a $\cos^2\theta$ angular dependence. Similar angular dependence has been reported in the in-plane AMR of manganites such as $La_{1-x}Sr_xMnO_3$.[25] However, in the low temperature COO state ($T < T_{COO}$), the AMR deviates from the $\cos^2\theta$ dependence and there is an irreversible resistance drop accompanying the rotation of the magnetic field. The AMR shows different behavior near MIT and in the COO phase, suggesting strong correlation between AMR and the electronic phases in perovskite manganites.

Possible contribution of the sample geometry to the observed AMR effect has been examined. Both cuboid- and cylinder-shaped samples have been made from the sintered $Nd_{0.5}Sr_{0.5}MnO_3$ polycrystals [Figs. 2(a) and 2(b)]. From the comparison between AMR in the cuboid- and cylinder-shaped sample (Fig. 2), it is found that for $T > T_{COO}$ [Figs. 2(c) and 2(d)], the AMR in the cuboid-shaped sample is almost ten times larger than that in the cylinder-shaped one. Considering our measurement configuration, the cuboid-shaped sample has much larger shape anisotropy than the cylinder-shaped sample, thus it is reasonable to attribute the AMR to the shape anisotropy for $T > T_{COO}$. However, for $T < T_{COO}$ [Figs. 2(e) and 2(f)], AMR with comparable magnitude has been observed for both cuboid- and cylinder-shaped samples, indicating that the AMR cannot be contributed by shape anisotropy only. In order to avoid possible contribution of texture, we have cut a slab directly from the cylinder-shaped sample and measured the AMR effect. Similar results with that of the cuboid-shaped sample were obtained (not shown), indicating new mechanisms other than shape anisotropy are involved.

The unusual AMR effect is characterized by the irreversible resistance drop upon rotation of the magnetic field. In manganites with phase separation, the time relaxation effect may contributed to a continuously drop of the resistivity, i.e., after applying a constant magnetic field, the resistance may slowly change with time.[26] Figure 3(a) shows the relaxation effect at 80 K under different magnetic fields along fixed direction. It is found that the resistance changes were less than 4% after 10 min, indicating that the relaxation effect only contributes to a small portion of the observed AMR in $Nd_{0.5}Sr_{0.5}MnO_3$ polycrystals. We have also conducted the AMR measurement after waiting for long enough time (30 min) with $H$ applied at $\theta = 0$ (not shown). Considerable resistance drop upon rotating $H$ was still observed, manifesting that the time relaxation effect cannot be accounted for the unusual AMR effect.

It is observed that the unusual AMR strongly depends on the applied magnetic field strength [Fig. 2(e)]. At low field (such as 0.5 T), only a small resistance drop is observed. With increasing magnetic field, the irreversible resistance

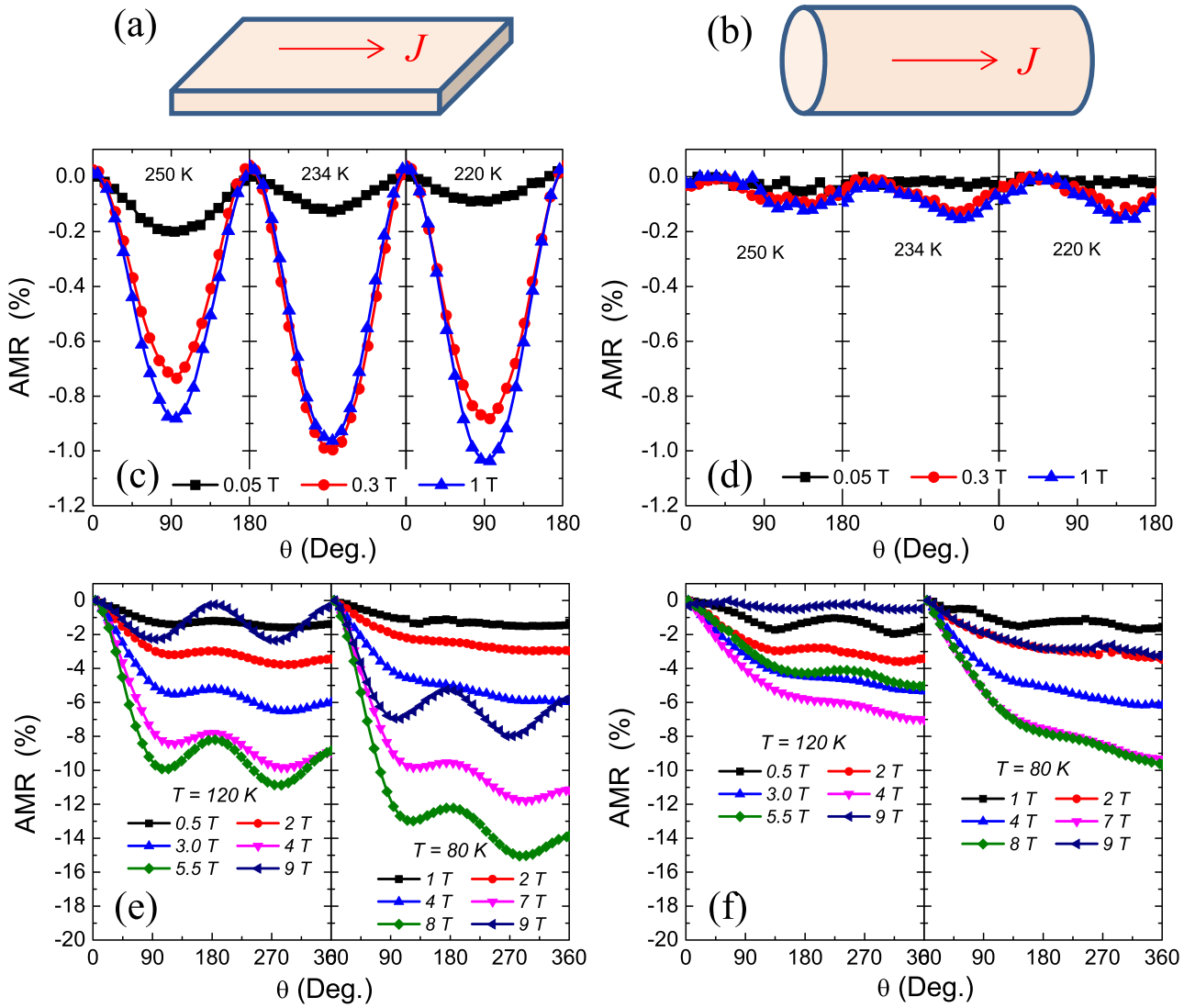


FIG. 2. (a) and (b) Schematic picture showing the geometric shape of the cuboid- and cylinder-shaped samples. (c) and (d) Comparison of AMR near MIT in the cuboid- and cylinder-shaped samples. (e) and (f) Comparison of AMR in the COO state in the cuboid- and cylinder-shaped samples.

drop dominates the AMR ($H = 2$ T). At larger magnetic field ($H > 4$ T), there is again negligible irreversible resistance drop in AMR, as the COO state has been transformed into the FMM state [Fig. 3(b)].

In order to understand the origin of the unusual AMR, we have subtracted the shape anisotropy and relaxation effect from the experimental curve. Figure 3(c) shows the result at 80 K under 6 T field. It is clear that after subtraction, the remaining AMR shows nearly linearly drops of the resistance when $\theta$ is small and saturates when $\theta$ approaches 180°. In the out-of-plane AMR configuration, the relative orientation between $M$ and $I$ is kept constant, thus explanation similar to the in-plane AMR fails. Recent reports has shown similar angular dependent AMR in epitaxially grown manganite thin films, the origin of which is believed to correlates with the phase competition.[21–23] In our cases, we suggest the phase competition between COO phase and the field-induced FMM phase may also contribute to the observed unusual AMR effect. Furthermore, the unusual AMR [Fig. 4(d)] shows its peak value at intermediate field strength and vanishes at high magnetic fields where the COO has been turned into FMM state, indicating a major role played by phase competition.

The correlation between phase competition and the unusual AMR has been understood by considering the expansion of the volume of FMM domains due to the rotation of FMM domains with preferential expansion along the field direction.[23] However, this postulation has neglected the fact that there is strong crystalline orientation dependent magneto-transportation behavior, which has been observed in perovskite manganite single crystals.[19,20] Based on this fact and by considering all the observations, we propose the following model: (1) the sample resistance depends mainly on the volume fraction ($f_{COO}$) of the COO state

$$R = R(f_{COO}(T, \vec{H})) \tag{1}$$

and the magnetoresistance is caused by the field induced melting of the COO domains. (2) For a single COO domain, the energy barrier for field-induced COO melting depends not only on the magnitude of magnetic field but also on the orientation. As we know, $Nd_{0.5}Sr_{0.5}MnO_3$ exhibits

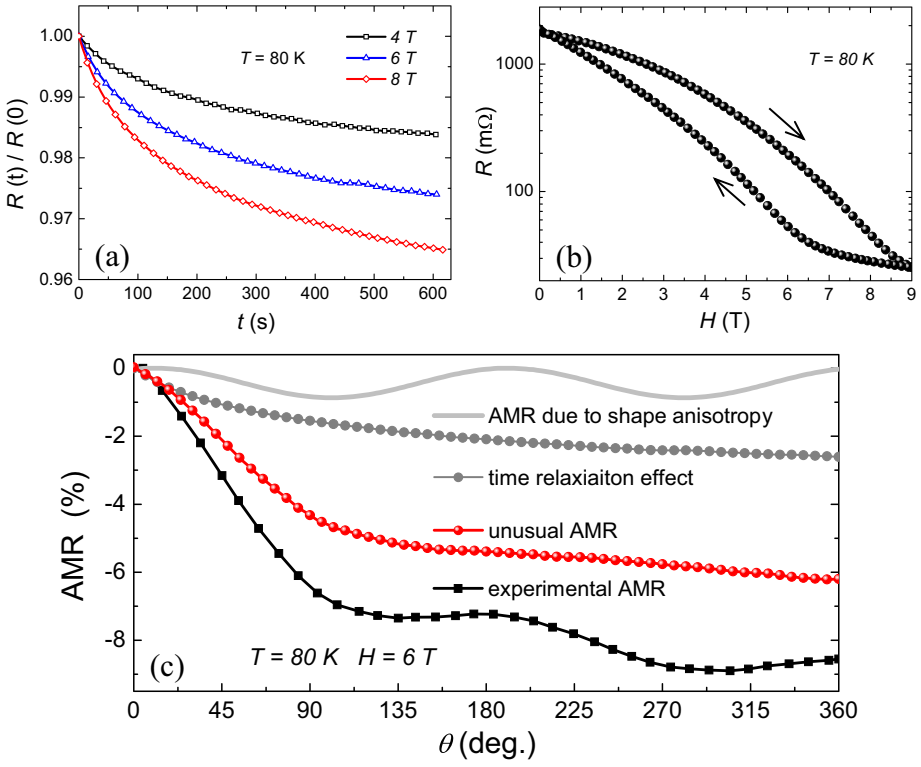


FIG. 3. (a) Resistance decaying at 80 K with applied magnetic fields of 4, 6, and 8 T, respectively. The time is approximately the same to that for magnetic field rotate from $\theta = 0°$ to $\theta = 360°$. (b) Magnetic field dependence of resistance for $Nd_{0.5}Sr_{0.5}MnO_3$ at 80 K (COO state), the arrows indicate the field ramping directions. (c) Fitting of the measured AMR curve at 80 K and 6 T reveals three contributions: demagnetization effect, time relaxation effect, and an unusual AMR effect.

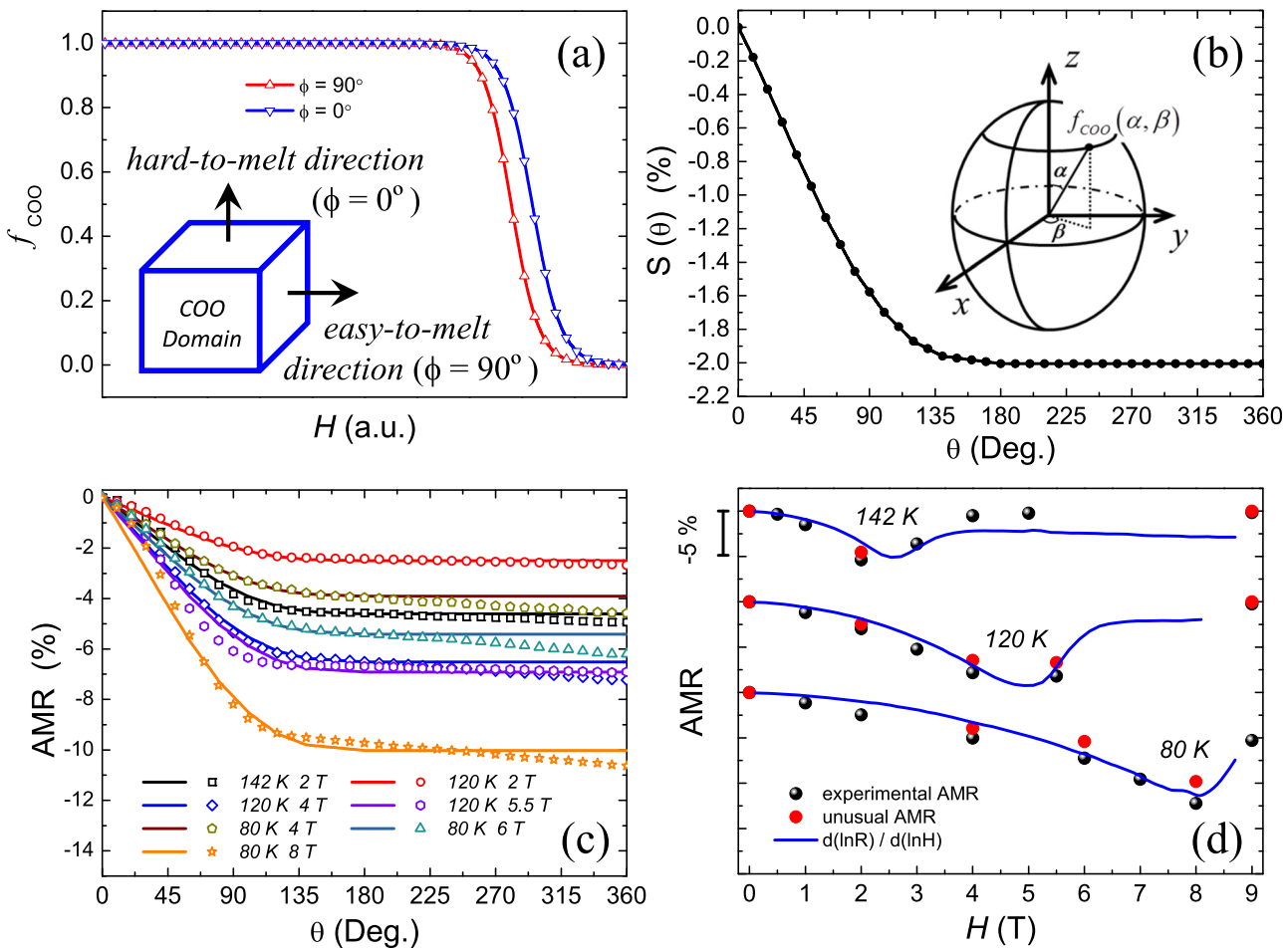


FIG. 4. (a) Schematic showing of anisotropic magnetic field induced melting of COO domains, the $f_{COO}$ is larger for $\phi = 0°$ than $\phi = 90°$. (b) Angular dependence of $S(\theta)$. Inset shows the total COO volume fraction, which can be represented by an ellipsoid when the magnetic field is applied along $\theta = 0°$ ($z$ axis). (c) Fitting of the AMR curves according to Eq. (4). (d) Magnetic field dependence of the unusual AMR at 142 K, 120 K, and 80 K, the field dependence of the experimental AMR value has been plotted as guidance, and the AMR value predicted by the model is also shown. The data have been offset for clarity.

charge-exchange (CE) type ordering in the COO state. In a COO domain, the spins and orbitals lie in the *ab*-plane that is extended along the *c*-axis.[27] As a result of spin-orbital coupling, the energy barrier between the COO and FMM states should depend on the magnetic field orientations. Take $\phi$ as the angle between the magnetic field and the hard-to-melt direction (then $\phi = 90^\circ$ correspond to the easy-to-melt direction), to first order simplification, we may write

$$f_{COO}(T,\vec{H}) \approx f_{COO}(T, H(1+\lambda\phi)), \tag{2}$$

where $\lambda$ is a coefficient that describes the strength of the anisotropy. At $H = 0$, the anisotropy disappears naturally. Figure 4(a) schematically shows the anisotropic field dependence of $f_{COO}$ for well-ordered COO domains. Due to the anisotropic melting, $f_{COO}$ is larger for $\phi = 0^\circ$ than that with $\phi = 90^\circ$. Assuming the anisotropy is weak ($\lambda\phi \ll 1$),[28] then a near linear decrease of the COO fraction with varying $H$ from $\phi = 0^\circ$ to $\phi = 90^\circ$ will be expected, and the AMR may be expressed as

$$\begin{aligned} AMR(\phi) &= \frac{\Delta R}{R} \approx \frac{1}{R}\frac{dR}{df_{COO}^{\phi}} \cdot \frac{df_{COO}^{\phi}}{dH} \cdot \lambda H\phi \\ &\approx \frac{d(\ln R)}{d(\ln H)} \cdot \lambda\phi. \end{aligned} \tag{3}$$

For the above discussion, we have only considered well-ordered COO domains. In polycrystalline sample, the COO domains are randomly orientated. When the magnetic field is applied along $\theta = 0$ ($z$ axis), the total COO volume fraction is represented by an ellipsoid, with $f_{COO}(\alpha, \beta) = f_{COO}(\alpha)$ corresponds to COO volume fraction of domains with their easy-to-melt direction parallel to ($\alpha$, $\beta$), as shown in the lower inset of Fig. 4(b). Considering that the transition from COO to FMM state is of first order, we expect $f_{COO}$ to continuously decrease with $\theta$. The corresponding AMR may be expressed as

$$AMR(\theta) = \frac{\Delta R}{R} \approx \frac{d(\ln R)}{d(\ln H)} \cdot S(\theta). \tag{4}$$

The factor $S(\theta)$ can be obtained by using graphical method[29] as plotted in Fig. 4(b). It is found that $S(\theta)$ first decrease linearly with $\theta$, then goes into saturation when $\theta$ is increased up to 180°. We have fitted the unusual AMR according to Eq. (4). As shown in Fig. 4(c), the fitting results agree with the unusual AMR. The field dependence of AMR value, as suggested from Eq. (4), varies as $\mathrm{d}(\ln R)/\mathrm{d}(\ln H)$. Fig. 4(d) depicts the fitting of the magnetic field dependence of the AMR value at typical temperatures below $T_{COO}$. The fitting generally agrees with the experimental results, and the derivations may be due to the fact that the magnetoresistance is not solely induced by the melting of COO state. The magnetoresistance in the FMM state also contributes, as shown in Fig. 3(b).

## IV. CONCLUSION

We have studied the out-of-plane AMR effect in charge-orbital ordered $Nd_{0.5}Sr_{0.5}MnO_3$ polycrystals. It is observed that for $T > T_{COO}$, the AMR is mainly induced by the shape anisotropy. However, in the COO state ($T < T_{COO}$), there is an unusual AMR that shows irreversible resistance drop upon rotation of the magnetic field. The unusual AMR correlates with phase competition between the COO phase and the field induced FMM phase, reaches a value as large as 10% even in polycrystals. A model based on anisotropic magnetic field induced melting of the COO domains has been proposed, which describes both the angular dependence and magnetic field dependence of the AMR very well.

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