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# Giant anisotropic magnetoresistance in bilayered $La_{1.2}Sr_{1.8}Mn_2O_7$ single crystals

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We report an observation of anomalous anisotropic magnetoresistance (AMR) in bilayered $La_{1.2}Sr_{1.8}Mn_2O_7$ single crystals. A giant AMR is found to be 80% under a magnetic field of 1 T near the metal-insulator transition temperature, where AMR is defined as $\text{AMR}=[\rho(H\perp \boldsymbol{c})-\rho(H\|\boldsymbol{c})]/\rho(H\perp \boldsymbol{c})\times 100\%$, and $\rho(H\perp \boldsymbol{c})$ and $\rho(H\|\boldsymbol{c})$ are the resistivity with the magnetic field perpendicular and parallel to $c$-axis, respectively. The AMR effect shows strong temperature and magnetic field dependences, and indicates a close interrelation with the anisotropic field-tuned metal-insulator transition. © *2011 American Institute of Physics*. [doi:10.1063/1.3593486]

Anisotropic magnetoresistance (AMR), the property of resistivity varying with the magnetic field orientation with respect to either the electrical current or the crystal axes, has been studied widely in magnetic materials.[1–10] The dependence of resistivity on the field direction relative to the electrical current is expected due to the Lorentz force acting on the conduction electrons while the dependence of resistivity on the field orientation with respect to the crystal axes is related to the spin-orbital and magnetoelastic couplings.[6,7] The study of AMR can provide detailed information about the underlying physics, and also have wide technological applications. Recently, it was found that the AMR indicates some extraordinary behaviors in perovskite manganites.[2–10] For example, in manganite thin films, the AMR effect shows a magnitude of more than 30%, and exhibits strong and nonmonotonic temperature ($T$) and field ($H$) dependences, which are dramatically different from that in the conventional ferromagnetic metals or alloys.[2–9] In single crystal $La_{0.69}Ca_{0.31}MnO_3$ with a pseudocubic structure and a weak anisotropy, Li *et al.*[10] reported a large AMR effect of about 90% under an magnetic field of 0.2 T near the metal-insulator transition (MIT) temperature ($T_{MI}$), which shows a closely relation with the anisotropic MIT. At present, the study of the AMR effect in bilayered manganites $La_{2-2x}Sr_{1+2x}Mn_2O_7$, in which a stronger anisotropy is expected, has not been performed. The $La_{2-2x}Sr_{1+2x}Mn_2O_7$ crystal is composed of $MnO_2$ bilayers which are stacked along the $c$ axis and separated by insulating nonmagnetic $(La,Sr)_2O_2$ layers, leading to a quasi-two-dimensional (2D) structure.[11] Due to the reduced dimensionality, the crystal shows strong crystalline anisotropy and anisotropic magnetotransport behavior,[12,13] and thus large AMR effect should be expected. In this letter, we report detailed studies of AMR effect with respect to the electrical current and crystal axes in $La_{1.2}Sr_{1.8}Mn_2O_7$ single crystals. A remarkable $T$- and $H$-dependent AMR effect was found, which shows a close interrelation with the anisotropic MIT in the system. We attribute the observed results to the unique magnetoelastic phenomena and strong spin-lattice coupling in the crystal.

A single crystal of $La_{1.2}Sr_{1.8}Mn_2O_7$ was grown by the floating-zone method in an optical image furnace. X-ray diffraction measurements on the powders show a pure perovskite phase. The composition of the crystal is checked by inductively coupled plasma atomic emission spectroscopy. The crystal was oriented using Laue x-ray diffraction patterns and cut into a rectangular shape with a size of $3\times 1.5\times 0.5\ \text{mm}^3$. The largest plane is the easy plane ($ab$ plane). The dc susceptibility was measured using a superconducting quantum interference device magnetometer (Quantum Design, SQUID). The MR measurements were carried out using a physical property measuring system (Quantum Design) equipped with a motorized sample rotator. The standard four-probe method was used for the resistivity measurements. The angular dependence of the resistivity $\rho(\Theta)$ was measured at a fixed magnetic field and temperature by varying the sample orientation relative to the field, where $\Theta$ is the angle between the field and $c$-axis in the plane perpendicular to the current direction.

Figure 1 shows the magnetization and resistivity as a function of temperature of the crystal. Resistivity is measured at a pulsed current of 100 $\mu$A and magnetization is measured in an applied field of 100 Oe. Well consistent with the reported results, a sharp ferromagnetic to paramagnetic transition occurs around 120 K.[13–15] Large anisotropy of the low-field magnetization indicates that the easy plane lies in the $MnO_2$ layer. Around 120 K, a MIT is observed with the sharp drop of resistivity by more than two orders of magnitude. Thus, here we define the temperature where the resistivity begins to drop [seen the inset of Fig. 1(b)] as the $T_{MI}$. The measured $\rho(T)$ versus $H$ curve, shown in Fig. 1(b), demonstrates a typical negative MR behavior.

Figure 2 shows the angle and temperature dependences of the normalized resistivity $\rho(T,\Theta)$ measured under a mag-

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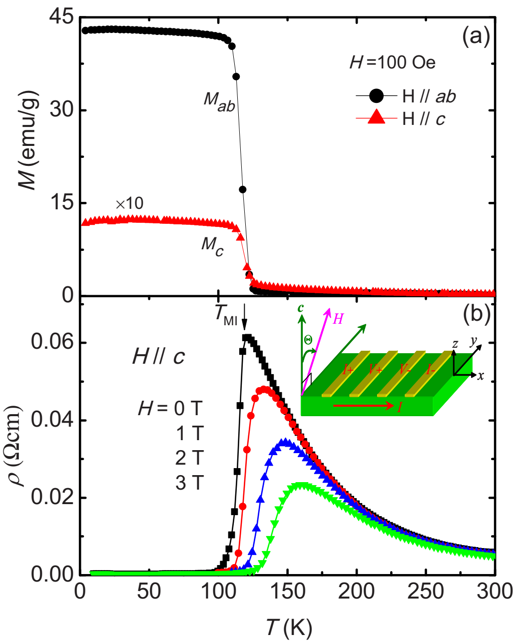


FIG. 1. (Color online) (a) Magnetization and (b) resistivity as a function of temperature of the $La_{2-2x}Sr_{1+2x}Mn_2O_7$ ($x$=0.4) single crystal. The inset of (b) shows the schematic for the resistivity and AMR measurements.

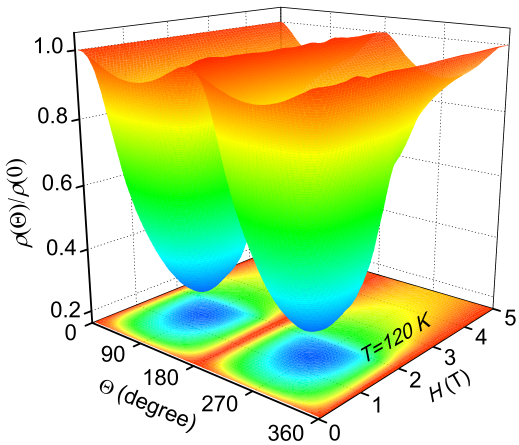


FIG. 3. (Color online) Angle and $H$ dependences of the normalized resistivity, measured with external magnetic field rotating about the current direction at 120 K.

netic field $H$=1.0 T. In the measurement, the field $H$ rotates in the plane perpendicular to the current direction ($yz$ plane) with angle $\Theta$ between the field and $c$-axis [shown in the inset of Fig. 1(b)], and thus the conventional Lorentzian MR could be neglectable. The results demonstrate a strong $T$-dependent AMR effect with a peak close to $T_{MI}$. The AMR amplitude is defined as $R_p=[\rho(H\perp \mathbf{c})-\rho(H\| c)]/\rho(H\perp \mathbf{c})\times 100\%$, where $\rho(H\|\mathbf{c})=\rho(\Theta=0)$ and $\rho(H\perp \mathbf{c})=\rho(\Theta=90°)$ are the resistivities for a given field being parallel and perpendicular to the $c$-axis of the crystal, respectively. For $H$=1.0 T, a peak value of $R_p\sim 80\%$ is observed near $T_{MI}$. The large AMR appears only near $T_{MI}$, not in either the pure ferromagnetic metallic state or the pure paramagnetic insulating state.

Figure 3 presents the normalized resistivity $\rho(H,\Theta)$ measured at 120 K, which demonstrates a nonmonotonic field dependence with a peak near 1.5 T and the peak value is $\sim$80%. As further increasing $H$, the value of $R_p$ reduces gradually and only 4% left as $H$ approaches to 5 T. One can anticipate that the AMR effect would disappear as the field becomes strong enough. At other temperatures near $T_{MI}$, the AMR effect shows similar field dependence though the largest $R_p$ saturated appears at different field. By analogy with $La_{1-x}Ca_xMnO_3$ oxides,[10] such anomalous $H$-dependent AMR effect, as well as the $T$-dependent behavior shown in Fig. 2, should be related to the magnetocrystalline anisotropy and anisotropic MIT in the crystal.

To verify the above idea, the field dependence of MIT was investigated. As shown in Fig. 4, at a given field, the $\rho(T)$ curves show a clear shift when varying the magnetic field direction from $ab$ plane to $c$-axis, indicating an anisotropic $H$-dependent MIT. As presented in the upper inset of Fig. 4, the field dependence of $T_{MI}$ for the $H\|c$-axis deviates from that for the $H\|ab$ plane. In addition, the gap ($\Delta T_{MI}$) between two directions shows a nonmonotonic field dependence (shown in the lower inset of Fig. 4). Here, $\Delta T_{MI}$ is defined as $\Delta T_{MI}=T_{MI}(c)-T_{MI}(ab)$, where $T_{MI}(c)$ and $T_{MI}(ab)$ are the $T_{MI}$ with $H$ along $c$-axis and in $ab$ plane, respectively. $\Delta T_{MI}$ reaches the maximum near 3 T and then begins to decrease as field increases gradually. It can be expected that $\Delta T_{MI}$ will decrease further to zero, and reflecting the vanishing of anisotropy of $H$-dependent MIT under a sufficiently strong field. Comparing the results shown above, one may notice a close interrelation between the AMR effect

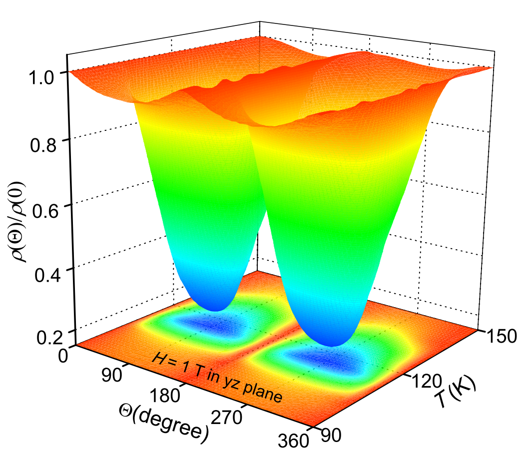


FIG. 2. (Color online) Angle and $T$ dependences of the normalized resistivity, measured under an $H$=1 T magnetic field rotating about the current direction.

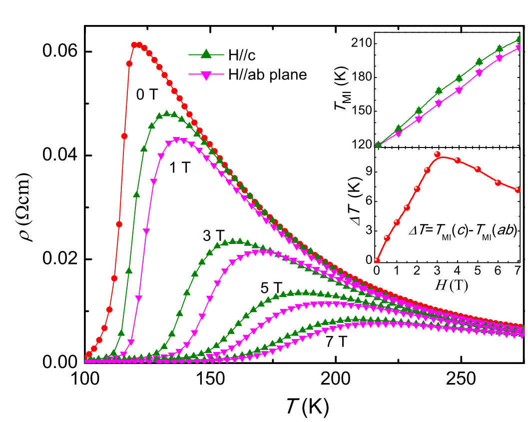


FIG. 4. (Color online) $T$ dependence of resistivity under different magnetic field along $c$-axis and $ab$ plane. The upper inset presents the $T_{MI}$ measured along two directions. The lower inset shows the difference $\Delta T_{MI}$ of $T_{MI}$ along two directions. The error bars are smaller than the symbols.

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and MIT. At a given field and temperature, the sample remains metallic when the field in *ab* plane while becoming insulating when the field is reoriented along *c*-axis. The difference in $T_{MI}$ due to field orientations leads to the observed unusual AMR effect. When the system is far away from the gap region of the two $T_{MI}(H)$ curves, only conventional AMR effect with a small amplitude exist.

To understand the anisotropic magnetotransport behavior, we should take account of the anisotropic crystal structure and magnetoelastic behavior. In bilayered manganites, the Mn–O–Mn network separated by insulating $(La,Sr)_2O_2$ layer along *c*-axis results in a 2D tetragonal structure ($I4/mmm$). As compared to the orthorhombic $ABO_3$ with a three-dimensional network of $MnO_6$ octahedra, these layered manganites have a reduced exchange coupling between the Mn ions along the *c* direction. In the studied $La_{1.2}Sr_{1.8}Mn_2O_7$, the Mn spins are ferromagnetically coupled within the $MnO_2$ bilayers[16] with an easy axis in *ab* plane [as shown in Fig. 1(a)]. With applying an external field near the paramagnetic-to-ferromagnetic phase transition as well as MIT, the alignment of Mn spins in *ab* plane is increased, thus leads to an enhancement of spin-polarized conductivity of $e_g$ electrons in $MnO_2$ sheets. This conductivity enhancement in turn results in a contraction of the planar Mn–O bonds in response to localized charge moving out of Mn–O antibonding orbitals to become carriers.[14] The contraction of planar Mn–O bonds leads to lattice shrinking along *ab* plane while expanding along *c*-axis.[14,15] Due to the easy axis lying in *ab* plane, the application of a magnetic field in *ab* plane can influence the spin alignment and lattice distortion (shrinking in *ab* plane and expanding in *c*-axis) more efficiently compared to applying the magnetic field alone *c*-axis (the hard axis). In other words, the structural anisotropy leads to the anisotropic lattice as well as spin response to the external field, and an anisotropic MIT tuned by magnetic field. We should note that the strength of structural anisotropy is not related to the AMR value directly. For $La_{0.31}Ca_{0.69}MnO_3$ single crystal, the structural anisotropy is much weaker than that in $La_{1.2}Sr_{1.8}Mn_2O_7$ but an AMR effect of 90% was observed. In addition, the twinning might exist in the crystal which is hard to avoid in perovskite manganites. The AMR value could be reduced somehow due to the twining effects.[10]

In summary, the AMR effect of bilayered $La_{1.2}Sr_{1.8}Mn_2O_7$ single crystals was studied. A giant AMR of ~80% was found near $T_{MI}$ under a magnetic field of 1 T. The AMR shows a strong temperature and magnetic field dependence with respect to the crystal axes. The observed anisotropic magnetotransport behaviors are attributed to the anisotropic lattice strain tuned by magnetic field in this layered system.

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