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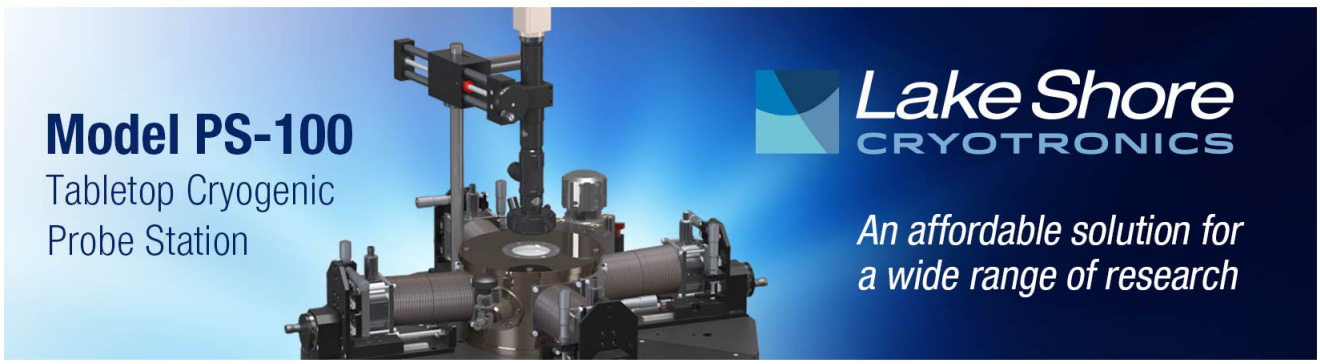

Correlation between the ferromagnetic metal percolation and the sign evolution of angular dependent magnetoresistance in $Pr_{0.7}Ca_{0.3}MnO_3$ film

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Angular dependent magnetoresistance (AMR) phenomena in $Pr_{0.7}Ca_{0.3}MnO_3$ film have been investigated. A transition between $\cos^2\theta$ dependent AMR in an insulating state at low fields and $\sin^2\theta$ dependent AMR in a metal state at high fields is observed at intermediate fields, depending on the temperature and/or strength of an applied magnetic field. Although the AMR sign evolution process from $\cos^2\theta$ dependence to $\sin^2\theta$ dependence at low temperature is different from that at high temperature due to existence of ferromagnetic insulator besides charge ordering antiferromagnetic insulator, we believe that such AMR sign evolutions are closely related with magnetic-field-induced ferromagnetic metal percolation behavior. © *2011 American Institute of Physics*. [doi:10.1063/1.3670399]

A great deal of research has been focusing on the anisotropic magnetoresistance or angular dependent magnetoresistance (AMR) since its discovery, particularly, for its potential applications in magnetic data storage. Recently, AMR has been reported in different colossal magnetoresistant (CMR) manganites.[1–4] For example, AMR was observed in the ferromagnetic manganites and shows a peak close to its Curie temperature with a non-monotonic temperature or magnetic-field dependence. This is very different from AMR in conventional ferromagnetic alloys. AMR was also observed in charge ordering $Sm_{0.5}Ca_{0.5}MnO_3$ films and coexistent-phase $La_{0.67}Ca_{0.33}MnO_3$ films. It has been noted that these AMR phenomena are closely connected with the transition from an antiferromagnetic insulator (AFI) to a ferromagnetic metal (FM), similar to CMR phenomena.

Bulk $Pr_{0.7}Ca_{0.3}MnO_3$ (PCMO) has been a very interesting system because of its low temperature physical properties. The low-temperature ground state (below 110 K) is the microscopically inhomogeneous coexistence of a charge-ordering antiferromagnetic insulator (CO-AFI) phase and a ferromagnetic insulator (FI) phase. Moreover, with a decrease of temperature, the quantities of FI fraction increase. The ferromagnetism in FM is well understood to be through a double exchange interaction, while the mechanism behind the ferromagnetism in FI is still not clear. Most importantly is that the CMR effect in most manganite oxides is thought to be closely related to insulator-metal transition through percolation,[5,6] which implies a continuous transformation, but it was reported in bulk PCMO that the evolution of the spin-wave stiffness at the CMR transition was first order and consequently not consistent with percolation.[7] Despite this contradiction, it was later proposed[8] that a percolative transition can be a first-order transition blurred by disorder or inhomogeneities. Last year, Saurel *et al.*[9] carried out a magnetic small-angle neutron scattering study on PCMO crystals. Their results confirmed that, with the application of a magnetic field, the magnetic transformation associated with the CMR effect has been found to occur by nucleation and growth of mesoscopic FM regions. They have shown that the insulator-metal transition is induced by the percolation of these FM regions.

In this letter, we studied AMR phenomena in nearly unstrained PCMO films to further confirm or understand the phase separation related to percolation. It has been found that the AMR sign evolution process from $\cos^2\theta$ dependence to $\sin^2\theta$ dependence is closely related with magnetic-field-induced FM percolation behavior.

80 nm PCMO films were deposited on $SrLaGaO_4$ (SLGO) substrate by sputtering methods. Details of the optimal deposition conditions can be found in Ref. 10. Bulk PCMO first undergoes a charge ordering transition in the range of 200–250 K and then transforms to CE-type antiferromagnetic (AF) state in the range of 130–140 K followed by the appearance of ferromagnetic clusters at 110 K with a further decrease of temperature.[11] Bulk PCMO crystallizes in an orthorhombic structure with lattice constants $a = 5.426$ Å, $b = 7.679$ Å, and $c = 5.478$ Å. In terms of pseudocubic lattice constants, the a-c plane becomes $a = 3.854$ Å, and the b-axis is $b = 3.839$ Å, which closely matches the lattice constant of the SLGO substrate, $a = 3.843$ Å. The as-grown PCMO film is cubic with lattice constant of 3.8406 Å, which is very close to bulk. The AMR(θ) measurements were performed using a physical property measurement system equipped with sample rotators. The AMR(θ) measurements for our test conditions are as follows: both the magnetic field (**B**) and current (**I**) are in the a-c plane direction, and θ is the angle between **B** and **I** with 0° defined as $\mathbf{B}\perp\mathbf{I}$. The film was rotated from 0° to 360° while the resulting AMR was being recorded.

Figure 1 shows R-T curve for PCMO film in the a-c plane. It can be seen that the resistance increases monotonously with decreasing temperature. No signature of a charge-ordering temperature, T_{CO}, can be found, but we

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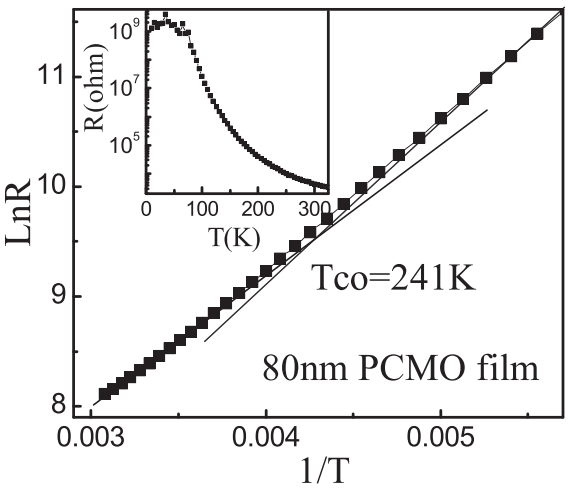

FIG. 1. lnR versus 1/T for PCMO film. Inset: R-T curve.

showed before that T_{CO} can be extracted by plotting lnR against 1/T.[12] The T_{CO} is found to be 241 K which is in the charge-ordering temperature range of bulk. It has been reported in our previous work[10] that unstrained PCMO films grown on SLGO behave quite similarly to bulk material. The R-T curves under different magnetic fields indicate that a transition from the insulating to the metallic state starts around 5 T below 50 K, and the metallic state is well developed at 7 T and 9 T at certain temperatures below 160 K.[10]

Figure 2(a) shows R-μ_0H curves for PCMO film at 75, 100, 125, and 150 K. At 125 K and 150 K, below T_{CO}, the resistance shows a continuous and slow decrease with increasing magnetic field, and little hysteresis is presented. However, at 75 K and 100 K, with magnetic field increasing, the resistance decreases rapidly with obvious hysteresis after a critical field of around 6 T is reached. Moreover, it is clearly seen that at 75 K, the resistance decreases by more than five orders of magnitude (compared to zero field) under a magnetic field of 9 T. It is also noted that at these four temperatures, the resistance does not show a tendency to saturate at the maximum applied field of 9 T. These results indicate that, at low temperatures (100 K and 75 K), a magnetic field of 6 T can induce a transition from an insulating to a metallic state, while at high temperatures (125 K and 150 K), one cannot conclude that such a transition occurs. In addition, it is found from the R-T curves under different magnetic fields in Ref. 10 that at 125 K (7 T) and at 150 K (9 T), the volume fraction of the magnetic field induced metallic phase is near the percolation threshold.

Below T_{CO} (=241 K), no obvious AMR(θ) is observed in our PCMO film until the temperature decreases to 150 K. This is different from what happens in the charge-ordered $Sm_{0.5}Ca_{0.5}MnO_3$ film[3] where the AMR(θ) oscillates under a magnetic field higher than 6 T once the charge ordering forms. Figures 3(a) and 3(b) represent the angular dependence of AMR at 150 K and 125 K under different magnetic fields, where $AMR(\theta) = [R(\theta) - R_{min}]/R_{min}$, and R_{min} is the minimum among the angular dependent resistances. At 150 K and 125 K, the AMR(θ) responses show a similar tendency of change with increasing magnetic field. AMR(θ) at 3 and 5 T shows $\cos^2\theta$ dependence with peaks at 0° and

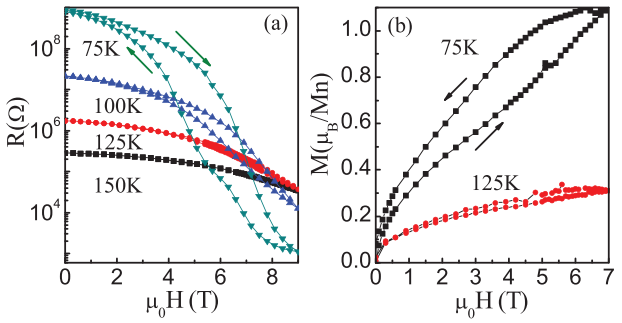

FIG. 2. (Color online) (a) R-μ_0H curves for PCMO film at 150, 125, 100, and 75 K. (b) M-μ_0H curves at 75 K and 125 K.

180° and valleys at 90° and 270°, albeit with a small deviation. With an increase of the applied magnetic field from 5 T to 9 T, the peak at 180° shifts towards higher angles and finally shifts to 270° at 9 T. Namely, AMR(θ) changes from $\cos^2\theta$ to $\sin^2\theta$ dependence gradually by increasing the field from 5 T to 9 T. The critical field for crossover from $\cos^2\theta$ to $\sin^2\theta$ dependence is 6.5 T at 125 K, while it is 8 T at 150 K. Crossover from $\cos^2\theta$ to $\sin^2\theta$ dependence was also observed in $Sm_{0.5}Ca_{0.5}MnO_3$ film[3] which exhibits a field-induced FM response but with no peak shifting being observed with an increasing magnetic field. Moreover, the sign crossover from $\cos^2\theta$ to $\sin^2\theta$ dependence is due to a collapse of charge-orbital order under 13 T as the metal state becomes very developed. Meanwhile, at 125 K and 7 T or at 150 K and 9 T, our films are in an only barely metal state or metallic percolation. It can be seen from Fig. 2(b) that the magnetic moment of our PCMO film at 125 K under a magnetic field of 7 T (maximum field) does not show a tendency to saturate, indicating our film is not in a uniform FM state. So, we believe that the gradual AMR sign evolution from $\cos^2\theta$ to $\sin^2\theta$ dependence may be very closely related with the FM percolation in the CO-AFI background.

As introduced above, the CO-AFI coexists with FI at a temperature below 110 K in bulk. To characterize the AMR at

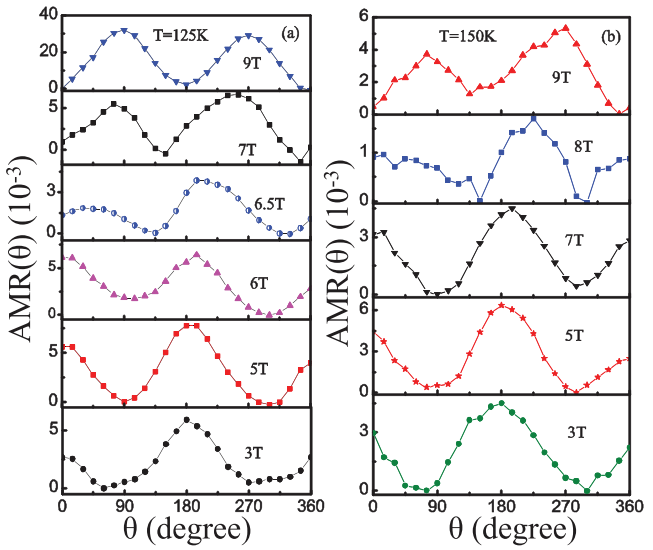

FIG. 3. (Color online) (a) and (b) Angular dependence of AMR(θ) at 150 K and 125 K under different magnetic fields.

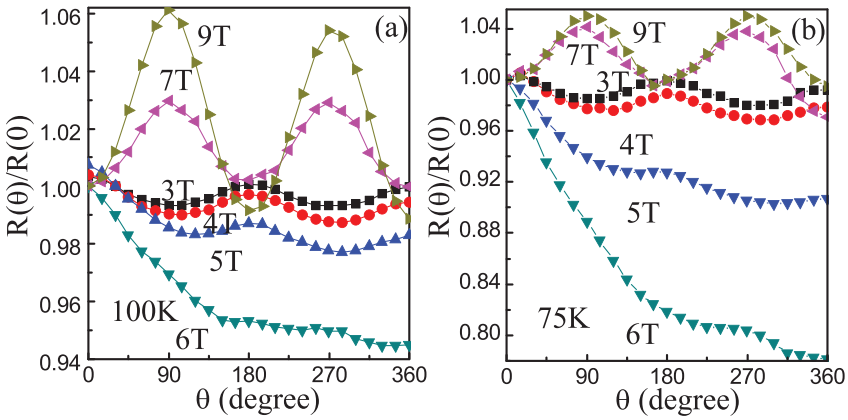

FIG. 4. (Color online) (a) and (b) Angular dependence of AMR(θ) at 100 K and 75 K under different magnetic fields.

100 K and 75 K, Figures 4(a) and 4(b) show the angular dependence of R(θ)/R(0) under different magnetic fields. It is found that at low magnetic fields (e.g., 3 or 4 T), R(θ)/R(0) still follows $\cos^2\theta$ dependence although the valley at 90° gradually shifts towards high angle with an increase of magnetic field from 3 T to 5 T. However, as the magnetic field becomes 5 T and/or 6 T at which point the magnetic field induced FM fraction is close to the percolation threshold (seen in Fig. 2(a)), R(θ)/R(0) drops quickly with a rotation of the magnetic field from 0° to 135° (and above) and keeps almost constant at higher angles, being more pronounced at 75 K than at 100 K. This is very different from what occurs at 125 K and 150 K (around the FM percolation threshold) due to the existence of a FI state along with the CO-AFI state. The mechanism behind the formation of FI domains is still not clear. When a magnetic field is applied, the transformation from CO-AFI into FM is clearly due to the magnetic energy between the magnetic field and the magnetic moments. In contrast, the same argument cannot be used for the transformation from FI phase into FM phase.[13] It was proposed in Ref. 13 that the interface between FM and FI is more strained than the FI to AFI interface. In order to reduce its lattice elastic energy, the system decreases the amount of FI as the FM phase is increasing. It was also reported in Ref. 9 that an insulator-metal transition takes place when the size of the FM phase reaches the percolation threshold, and the amount of FI starts to decrease. It is understood that around the percolation threshold, at 5 T or 6 T in our case, FI and FM phases start to compete, namely, the FM expands at the expense of the FI and also further at the expense of CO-AFI. During this process, we believe that disorder or inhomogeneity induced local ferromagnetic domain redistribution results in domain wall effects or different transformations. This occurs at the same magnetic field which leads to the above noted rapid drop of R(θ)/R(0) with the rotation angle increasing from 0 to 135° (and above). This phenomenon cannot be clearly explained here and requires further study. It was also reported in the AFI and FM phase-coexistent $La_{0.67}Ca_{0.33}MnO_3$ films[4] that $\rho(\phi)/\rho(0)$ follows a $\cos^2\theta$ dependence at a low magnetic field (0.5 T). When the magnetic field increases to 1.97 T, $\rho(\phi)/\rho(0)$ drops fast as the magnetic field rotates from 0° to 90° leading to larger AMR, and then it is suppressed at any other angle due to irreversible melting of AFI. However, the underlying reason is that different strain states along the a and b axes lead to a different $\rho(\phi)/\rho(0)$ response along these two axes. This was also reported in Ref. 6 where anisotropic strains make magnetic-field-induced FM filaments show directional ordering with preferential alignment along the [001] and [1-10] axes of the substrate. Because our PCMO film is almost unstrained, the effect of strain on AMR can be omitted in our case. Namely, the above explanation from Refs. 4 and 6 is not suitable for our system. In addition, it was also reported in the above $La_{0.67}Ca_{0.33}MnO_3$ film[4] that when the applied magnetic field is larger than the critical field, the films have a uniform FM state, and the AMR becomes negligible. However, in our case, under a magnetic field of 7 T and 9 T, the metallic state is well developed, with the AMR showing a $\sin^2\theta$ dependence. Furthermore, the magnitude of AMR increases with a magnetic field increase from 7 T to 9 T. It is seen from the M-H curve at 75 K (Fig. 2(b)) that a transition from both CO-AFI and FI to FM is continuous and with hysteresis. The magnetization at a maximum magnetic field of 7 T does not show a tendency to saturate, indicating that in our case, a magnetic field of 7 T cannot transform both CO-AFI and FI into a uniform FM state. So, the above insulator-metal transition occurring under a critical magnetic field of 6 T must be related to a percolation effect. With an increase of the magnetic field from 7 T to 9 T, more FM phases form leading to an increase of the AMR magnitude.

In conclusion, AMR phenomena including $\cos^2\theta$, $\sin^2\theta$-dependent AMR, and a transition between them have been observed in $Pr_{0.7}Ca_{0.3}MnO_3$ films, dependent on the temperature and the strength of the magnetic field. We believe that the AMR sign evolution from $\cos^2\theta$ dependence to $\sin^2\theta$-dependence is closely related with a magnetic-field induced FM percolation in the CO-AFI or phase coexistent CO-AFI and FI background.

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