Large low-field magnetoresistance of phase-separated single-crystalline $Pr_{0.7}Pb_{0.3}MnO_3$

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A large low-field magnetoresistance (MR) slightly above the metal–insulator transition temperature (234 K) was observed in single-crystalline $Pr_{0.7}Pb_{0.3}MnO_3$. Combining the temperature dependence of magnetization, resistance, and electron spin resonance spectra, it was suggested that phase separation occurs above the Curie temperature; ferromagnetic metallic clusters imbedding in the insulating paramagnetic matrix, and spin-polarized electron tunneling between isolated ferromagnetic clusters should be responsible for the large low-field MR observed. Undoubtedly, this observation opens a window to explore large low-field MR at high temperature, which is very important for the practical application of colossal MR effect.
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A great deal of attention has been focused recently on the colossal magnetoresistance (MR) effects in perovskite manganites with a formula $Ln_{1-x}A_xMnO_3$ (Ln=rare earth and A=alkaline earth elements).[1–4] In most cases, the large MR can be achieved only in a strong field in the Tesla range, which severely limits its application. Therefore, reducing the field scale and increasing the operating temperature have been the goal of a number of research groups worldwide. Hwang *et al.*[5] systemically investigated the MR of single-crystalline and polycrystalline $La_{2/3}Sr_{1/3}MnO_3$ in ferromagnetic (FM) metallic regime, and found that a large low-field MR appears in the polycrystalline sample at low temperature due to spin-polarized electron tunneling between grains, which does not occur in the single-crystalline sample, and the magnetic domain boundaries do not dominate the scattering process. Until now, there has been progress in reducing the field scale by exploring the extrinsic MR properties of polycrystalline samples in the form of thin films, bulk ceramics, and ultrafine powders.[5–14] These large low-field MR values achieved are very encouraging for practical applications. However, these large MR values are limited to low temperature and decrease rapidly with increasing temperature. As a result, how to obtain a large low-field MR at higher temperature becomes a more important issue. In this letter, we investigate systemically magnetic and transport behaviors of single-crystalline $Pr_{0.7}Pb_{0.3}MnO_3$, especially in the insulating regime slightly above the metal–insulator transition temperature (T_P). It was found that, when the temperature is slightly above the T_P, there still exist isolated FM clusters imbedded in the insulating paramagnetic (PM) matrix. The spin-dependent electron tunneling between isolated FM clusters can bring about a large low-field MR at higher temperature (235–245 K). Undoubtedly, this observation is very important for the future application of colossal MR effect.

The growth process of single-crystalline $Pr_{0.7}Pb_{0.3}MnO_3$ have been described in detail elsewhere.[15] The x-ray fluorescence result showed that the composition of the single crystal agreed within 2% of that expected from the chemical formula. X-ray diffraction with Cu $K\alpha$ radiation was used to examine the phase purity and the crystal structure. The result shows that the single crystal is of a cubic perovskite structure without any impurity phases. The magnetization measurements were performed in a commercial superconducting quantum interference device magnetometer. The micromagnetic properties were investigated by electron spin resonance (ESR) measurements carried out at 9.50 GHz using a

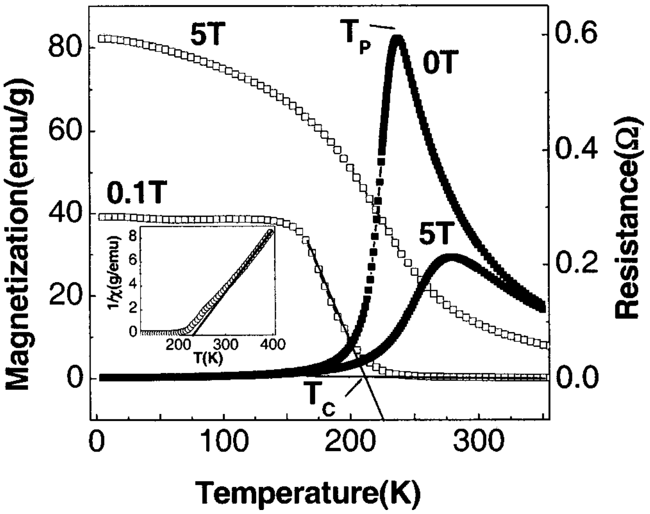

FIG. 1. Temperature dependence of magnetization (hollow symbol) and resistance (solid symbol) from 5 to 350 K measured in warm process under several magnetic fields. The definition of T_C and T_P are shown. The inset shows the inverse magnetic susceptibility, $1/\chi$, as a function of temperature. The straight line is a fit to the Curie–Weiss law.

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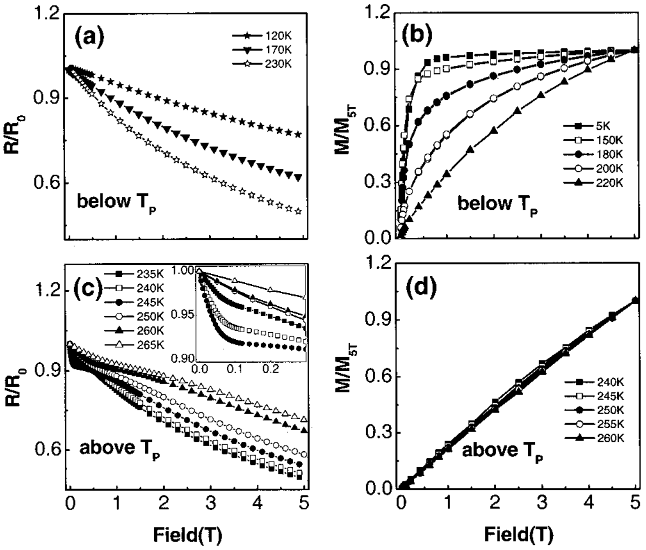

FIG. 2. Magnetic field dependence of the resistance normalized to the 0 T value (a) and (c) and magnetization normalized to the 5 T value (b) and (d) at various temperatures, panels *a* and *b* for the case below T_P, panels (c) and (d) for the case above T_P.

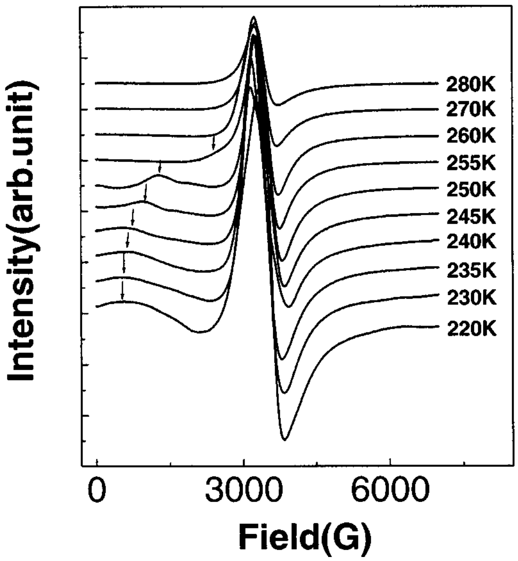

FIG. 3. ESR spectra measured in a field-sweeping mode (0–7000 G) at various temperatures.

BRUKER-200D spectrometer. The resistivity was measured by the standard four-probe method.

Figure 1 shows the temperature dependence of magnetization ($M-T$) and resistance ($R-T$) from 5 to 350 K measured in warm process under several magnetic fields. The sample shows a transition from FM to "PM" state at 210 and 278 K under a field of 0.1 and 5 T, respectively, and a metal–insulator transition at 234 and 280 K under a field of 0 and 5 T, respectively. It is noteworthy that the T_P, which is defined as the temperature corresponding to the resistance peak in $R-T$ curve, is nearly consistent with the Curie temperature (T_C) under a field of 5 T, however, is higher than the T_C under a lower field. In other words, under a low magnetic field, though long-range FM ordering is destroyed in the temperature range from the T_C to T_P, the sample can still keep metallic conductance.

Figure 2 shows the magnetic field dependence of normalized resistance [Figs. 2(a) and 2(c)] and magnetization [Figs. 2(b) and 2(d)] at various temperatures. When the temperature is below the T_P, the field dependent magnetization in Fig. 2(b) indicates FM characteristics up to the T_P though the temperature is above the T_C, and suggests that FM clusters with short-range FM interaction maybe exist above the T_C though the long-range FM ordering is destroyed, which is well consistent with the difference between the inverse magnetic susceptibility and its fit to Curie–Weiss law above T_C, as shown in the inset of Fig. 1. In the same time, there is an increasing negative MR smoothly varying through the magnetic field range studied, and the MR value at the same field increases with increasing temperature [see Fig. 2(a)]. Consistent with the results reported by Hwang *et al.*,[5] the electron scattering at magnetic domain boundaries seems not to be dominant in the transport process. The suppression of spin fluctuation should be the origin of the negative MR in this temperature range. It is noteworthy that, two striking features can be observed when the temperature is above the T_P: (i) the MR is radically different from the case below T_P, as shown in the inset of Fig. 2(c), a sharp drop in the resistance at low fields appears followed by a smooth decrease with increasing field, which is very similar to that observed in polycrystalline ceramics due to spin-polarized tunneling between grains. The low-field MR value at the same field increases up to 245 K ($\mathrm{MR}=9\%$ at 245 K under a field of 0.1 T with the definition $\mathrm{MR}=[R(0)-R(H)]/R(0)$, where $R(0)$ and $R(H)$ are the resistance at absence and presence of field), then disappears when the temperature is above 250 K. The high-field MR at the same field decreases with increasing temperature and (ii) as shown in Fig. 2(d), the sample indicates a linearly field-dependent magnetization, no obvious FM characteristic can be observed.

In order to shed further light on the micromagnetic properties of the sample near the T_P, the ESR spectra were measured in a field-sweeping mode (0–7000 G) from 220 to 280 K. As shown in Fig. 3, when the temperature is above 260 K, the ESR signals consist of a single peak with Lande factor $g=2.0$ independent nearly on the temperature. This signal has been believed to be due primarily to PM Mn ions.[16] Below 255 K, there is a clear extra FM resonance peak deviating from the resonance field position with $g=2.0$ and shifting to low fields with decreasing temperature. Obviously, in agreement with the magnetization measurements described herein, the ESR results indicate that, when the temperature is above the T_C, though the long-range FM ordering is destroyed, there still exist FM clusters imbedding in the PM matrix, which has been called phase separation. Similarly, FM clusters, whose size changes with temperature and field, imbedding in the PM matrix were also found by De Teresa *et al.*[17] and Goodenough and Zhou[18] in $La_{0.67}Ca_{0.33}MnO_3$.[17,18] When the temperature is between the T_C and T_P, FM clusters can provide continuous conducting path in the whole sample. As a result, the sample exhibits metallic behavior. Above the T_P, FM clusters become isolated, and the sample is transformed into a superparamagnetic state, therefore, no obvious FM characteristic can be observed in Fig. 2(d). Based on the analysis, aforementioned one can conclude that it is the spin-polarized tunneling between isolated FM metallic clusters that should be responsible for the large low-field MR observed slightly above the T_P.

In summary, in single-crystalline $Pr_{0.7}Pb_{0.3}MnO_3$, a large low-field MR was observed when the temperature is slightly above the T_P. The large low-field MR can be attributed to spin-polarized tunneling between isolated FM clusters imbedded in the insulating PM matrix. Though the low-field MR value is smaller than that observed by Hwang *et al.*[5] at low temperature, undoubtedly, this observation opens a window to explore large low-field MR at high temperature, which is very important for the practical application of colossal MR.

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