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Giant magnetoresistance and table-like magnetocaloric effect in double perovskite oxide $PrSrMnCoO_6$

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Magnetoresistance and magnetocaloric effect of a double perovskite oxide $PrSrMnCoO_6$ (cubic, $Fm\bar{3}m$) has been studied in fields up to 7 T. This compound is semiconductor-like and its electrical resistivity increases by 5 orders while going from 300 to 50 K. Giant magnetoresistance of ~40% is observed at 200 K in 7 T field. $PrSrMnCoO_6$ orders ferromagnetically at ~150 K and shows a maximum magnetic entropy change of ~4.6 J/kg/K for 5 T field change in the temperature range of 110–190 K. This nearly constant magnetocaloric effect over a broad temperature span is highly suitable for Ericsson-cycle magnetic refrigeration. © *2011 American Institute of Physics*. [doi:10.1063/1.3556716]

I. INTRODUCTION

The perovskite oxides are well known for their colossal magnetoresistance (CMR), multiferroic and superconducting properties.[1,2] Recently, double perovskite oxides of the general formula $AA'BB'O_6$ (A, A′—rare earth or alkaline earth ions, B, B′—transition metal ions) have attracted wide attention[3] as some of them are ferromagnetic at room temperature. The oxides such as Sr_2FeMoO_6 and La_2VRuO_6 exhibit large low field magnetoresistance at room temperature[4] and half-metallicity.[5] It is of substantial interest to look for materials with large magnetocaloric effect (MCE) near magnetic transition[6,7] because of prospective applications. Here we report giant magnetoresistance and moderate magnetocaloric effect over a broad temperature range in a double perovskite oxide $PrSrMnCoO_6$.

II. EXPERIMENTAL DETAILS

Polycrystalline samples of $PrSrMnCoO_6$ were prepared by a sol-gel method, the details of which are presented elsewhere.[8] The samples were characterized by x-ray powder diffraction, SEM and EDAX. The magnetization was measured using a superconducting quantum interference device magnetometer (MPMS, Quantum Design) and a vibrating sample magnetometer (PPMS, Quantum Design). Electrical resistivity has been measured by a standard four-probe method using PPMS.

III. RESULTS AND DISCUSSION

The powder x-ray diffraction data confirmed that the sample is single phase having cubic crystal structure (Space group $Fm\bar{3}m$) at room temperature with lattice parameter $a = 7.666$ Å. Figure 1 shows the magnetization (M) of $PrSrMnCoO_6$ in a magnetic field of 0.5 T after cooling in zero field (ZFC) and also after cooling in the measuring field (FC). The slope of the magnetization curve changes gradually in the temperature range of 50–200 K and the inflection point, 150 K, in the dM/dT curve is taken as the ferromagnetic transition temperature, T_C, of this sample. At temperatures below ~115 K, irreversibility between the ZFC and FC curves becomes noticeable, which is indicative of a magnetic glassy behavior.[9,10] The ZFC–FC magnetization separation could also result from anisotropy effects. The ZFC curve shows a broad maximum centered at 50 K and below this temperature the magnetization decreases with decreasing temperature, whereas the FC magnetization increases continuously down to 5 K. The inverse magnetic susceptibility in the temperature range of 200–300 K follows Curie–Weiss law and a paramagnetic Weiss temperature (θ_p) of 159 K and an effective magnetic moment (μ_{eff}) of ~6.12 μ_B/f.u. are obtained. At 5 K, the magnetization as a function of the applied magnetic field, $M(H)$ shows subtle hysteresis. The

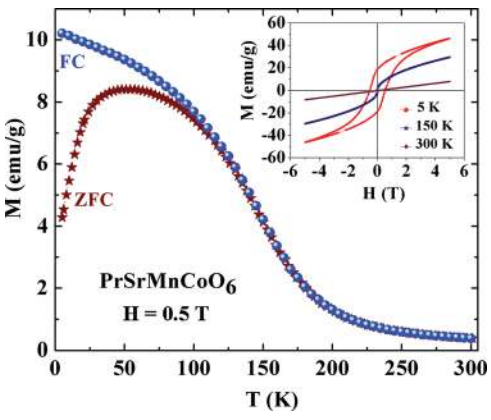

FIG. 1. (Color online) Temperature dependence of zero-field cooled and field cooled magnetization of $PrSrMnCoO_6$ in 0.5 T. (Inset) Magnetization vs field curves at three different temperatures.

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magnetization does not saturate in an applied field of 5 T [Fig. 1(inset)] and only a magnetic moment value of 1.63 μ_B/f.u. is obtained at 5 K in 5 T for this compound. It suggests possible ferrimagnetic alignment of moments. On the other hand, there is also a possibility of having some regions of Mn^{3+} and Co^{3+} moments in the lattice, which may give rise to local magnetic disorder[11] and hence the reduction in overall moment value and frequency dependence in ac magnetic susceptibility (see below).

The temperature dependence of the electrical resistivity of $PrSrMnCoO_6$ is shown in Fig. 2(a). The resistivity increases like a semiconductor from 300 to 50 K. The electrical resistivity value below 50 K is beyond the measurement limits of our instrument. In order to understand the nature of electronic conduction in the semiconducting region of the sample, we tried to fit the resistivity to Mott's variable range hopping (VRH) model $[\rho = \rho_0 \exp(T_0/T)^{1/4}]$[12,13] in the temperature range of 150–300 K. It is clear from the linear fits of ln ρ vs $T^{-1/4}$ [which is shown in Fig. 2(a, inset)] that the data fit well to the VRH-type of conduction.[14] $N(E)$ is calculated to be 3.28×10^{17} eV^{-1} cm^{-3} for the $PrSrMnCoO_6$ sample considering the value of L as 10^{-10} m (from Ref. 15). The value of $N(E)$ is of the same order of magnitude as in other known oxide semiconductors[16] [$N(E) \approx 10^{17}$–10^{19} eV^{-1} cm^{-3}]. Unlike most of the CMR manganites, the onset of ferromagnetic/ferrimagnetic order does not lead to metallic conductivity in $PrSrMnCoO_6$.

The magnetoresistance ratio (MR %) for $PrSrMnCoO_6$ as a function of applied field (H) at various temperatures is plotted in Fig. 2(b). The MR vs H plot shows a change in slope across the T_C. The compound shows giant MR of ~40% in an applied magnetic field of 7 T at 60 K. This MR value is very large compared to that of the double perovskite oxides, Sr_2CrMoO_6 (Ref. 17) and Sr_2FeMoO_6 (Ref. 18). The negative sign for MR obtained in the broad temperature range of 50–200 K suggests the dominant role of spin-dependent scattering in the resistivity of this compound. The site disorder at the B site does not seem to affect the magnetoresistance in this compound. The slope change in the MR% vs H graph at relatively high fields indicates quenching of spin fluctuations in large magnetic fields. We have also plotted MR vs M^2 at 100 K (in the magnetically ordered state) and find that it has linear dependence (figure not shown). This low temperature MR behavior is also observed in other ferromagnets[19] and could be attributed to the strain effects due to the domain alignment in applied field.

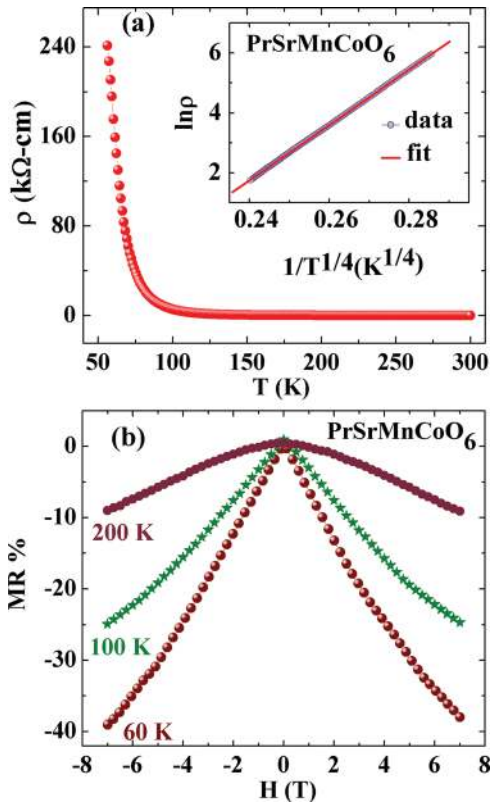

FIG. 2. (Color online) (a). Temperature variation of electrical resistivity, ρ, of $PrSrMnCoO_6$. (Inset) Fit to variable range hopping model. (b) Field dependence of magnetoresistance MR at different temperatures.

The magnetic entropy change in $PrSrMnCoO_6$ can be obtained by using the experimental isothermal magnetization data [Fig. 3(a)].[20,21,22] Figure 3(b) shows the temperature dependence of $|\Delta S_M|$ curves for magnetic field change (ΔH) from 0 to 5 T. The maximum change in magnetic entropy ($|\Delta S_M|_{max}$) is about ~4.6 J/kg/K for a field change of 5 T. The moderate value of $|\Delta S_M|_{max}$ is comparable to those observed in contemporary double perovskite oxides.[23] Apart from the large $|\Delta S_M|_{max}$, the relative cooling power (RCP) is an important parameter to determine the efficiency of magnetic cooling. RCP is defined as $RCP = |\Delta S_M|_{max} \delta T_{FWHM}$, where δT_{FWHM} is the full-width at half-maximum of $|\Delta S_M|$ vs T curve.[21] The value of RCP obtained is about 257 J/kg for 5 T, which is large compared to that in the other known CMR manganites.[21,24] The $|\Delta S_M|_{max}$ value is found to vary linearly with the change in magnetic field as plotted in the Fig. 3(c). This feature is preferred for efficient Ericsson-cycle-based magnetic refrigeration. The large value of MR%, MCE, and RCP compared to other double perovskites, easy preparation method, low processing cost of the materials compared to the rare earth intermetallic compounds, and high chemical stability make this compound a promising candidate for applications in magnetoresistive devices and magnetic refrigeration near transition temperature.

In order to confirm the glassy magnetic behavior suggested by the dc magnetization data, ac susceptibility has also been measured as a function of temperature at different frequencies (33, 133, 1333, and 9333 Hz) and is shown in Fig. 4. The peak temperature is found to shift to higher temperatures with increasing frequency following critical slowing down model.[25] In this model, the relaxation time τ is related to the correlation length ξ as $\tau \sim \xi^z$. As $\xi = [(T_f - T_g)/T_g]$, so $\tau/\tau_0 = [(T_f - T_g)/T_g]^{-z\nu}$, where $z\nu$ is the dynamical exponent, T_f is the freezing temperature, and T_g is the glass transition temperature. The fit to this model gives $z\nu = 4.8$ and $\tau_0 = 10^{-6}$s. This value of τ_0 is much longer than that of conventional spin glass ($\tau_0 \sim 10^{-13}$ s),[25] which indicates that the sample has magnetic clusters with weak intercluster interactions. It may give rise to the slow increase in magnetization near the magnetic transition and hence to a nearly constant MCE over a broad temperature range. The moderate

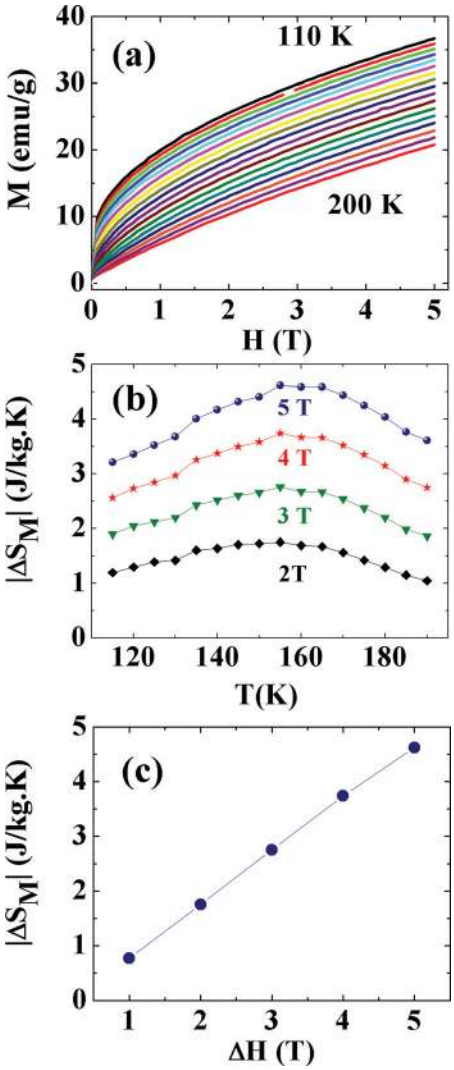

FIG. 3. (Color online) (a) Magnetic field dependence of the magnetization at selected temperatures in fields up to 5 T. (b) The magnetic entropy change $|\Delta S_M|$ as a function of temperature for various magnetic field changes. (c) $|\Delta S_M|$ as a function of the magnetic field change for $PrSrMnCoO_6$.

values of MCE realized in this compound could stem from the fact that the Co and Mn spins in the lattice may lead to noncollinear magnetic ordering and hence nonsaturation of magnetic moments even in large applied fields and some degree of frustration/competition that leads to the magnetic glassiness. The present study provides a key to design new double perovskite oxide materials by careful substitution to possibly obtain a first-order magnetic transition thereby achieve large magnetocaloric effect.

IV. CONCLUSIONS

In summary, the polycrystalline double perovskite oxide, $PrSrMnCoO_6$ is synthesized by a sol-gel method. This material orders ferromagnetically at $T_C = 150$ K. The maximum magnetoresistance is found to be ~40% at 60 K in applied field of 7 T. It also shows a moderate magnetocaloric

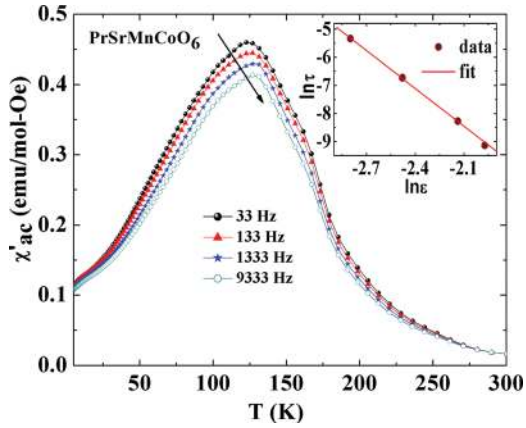

FIG. 4. (Color online) The temperature variation of the real part of ac susceptibility, χ' of $PrSrMnCoO_6$. (Inset) ln τ vs. ln ε data and linear fit.

effect near 150 K. The maximum entropy change at 155 K is about ~4.6 J/kg/K in a field change of 5 T.

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