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# Magnetoresistance and magnetostriction effects in bulk Dy-doped $La_{2/3}Sr_{1/3}MnO_3$

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ABSTRACT

Colossal magnetoresistance and large magnetostriction effects of bulk polycrystalline manganites $(La_{1-x}Dy_x)_{2/3}Sr_{1/3}MnO_3$ ($x = 0$–$0.4$) have been investigated in a magnetic field up to 50 kOe and in the temperature range 10–380 K. By increasing the dysprosium content $x$, the phase structure transits from a rhombohedral to an orthorhombic form at $x \sim 0.3$ due to the reduction of the average ionic radius of the A site $\langle r_A \rangle$, which results in not only the rapid decrease of Curie temperature and the enhancement of resistivity and magnetoresistance effect, but also the antiferromagnetic behavior at low temperature. Near the Curie temperature, the volume magnetostriction (VMS) of $x = 0.3$ sample reaches a maximal value about $-243 \times 10^{-6}$ in a magnetic field of 50 kOe. The large VMS effects can be attributed to the structural transition from an orthorhombic to a rhombohedral phase and the presence of a magnetic two-phase ferromagnetic-antiferromagnetic state.



## 1. Introduction

Since 1995 magnetostriction (MS) effects of manganites $R_{1-y}A_yMn_3$ (where R = La, Sm, Nd, Pr, Gd etc. and A = Sr, Ca, Ba, etc.) have been attracting considerable attention due to the close interplay among magnetotransport, magnetic and elastic properties [1–12]. The MS effects reported in the manganites generally possess two common features: (1), an obvious volume magnetostriction (VMS) effect for a manganite often occurs in the vicinity of the phase transition temperature, such as the Curie temperature ($T_C$) near $y \sim 0.33$ [1–9], charge ordering temperature ($T_{CO}$) near $y \sim 0.5$ [10–12]; (2), magnetic field and temperature dependences of MS exhibit an irreversibility as measured in both cooling and warming runs or in both increasing and decreasing magnetic field runs, which implies a first-order nature of the phase transition. So far several mechanisms of the MS effect in the manganites have been proposed, for example, the structural phase transition caused by applied magnetic field [1,2,5,12], the formation of small-radius polarons [6], the presence of a magnetic two-phase ferromagnetic–antiferromagnetic (FM–AFM) state due to strong s–d exchange [3,4,9], etc. Overall, the VMS behaviors of doped $La_{1-x}Sr_xMnO_3$ compounds were studied much less than those of doped $La_{1-x}Ca_xMnO_3$ compounds. Asamitsu et al. [1,2] studied the MS effect of the $La_{0.83}Sr_{0.17}MnO_3$ single crystal near $T_C$ due to the structure transition. Koroleva et al. [3,4] studied the giant VMS effect above 80 K of $La_{0.7}Sr_{0.3}MnO_3$ single crystal and found that the volume magnetostriction $\omega$ has a maximum of $\sim -50 \times 10^{-6}$ at 9 kOe near $T_C$. They attributed this phenomenon to the existence of a magnetic two-phase state consisting of a conducting FM matrix containing AFM insulating microregions due to a strong s–d exchange. However, the MS behaviors in bulk Dy-doped $La_{2/3}Sr_{1/3}MnO_3$ (LSMO) have not been reported up to now.

In perovskite-type manganites, the average ionic radius of A site $\langle r_A \rangle$ plays an important role in changing the crystalline structure, the bandwidth and magnetic and transport properties of mangnites [7,8,13–17]. Kundu et al. and Rao et al. had studied a series of manganites of the type of $(La - R)_{1-y}A_yMnO_3$ and found the occurrence of electronic phase separation due to chemical substitutions in the A-site [13–15], which resulted in magnetic inhomogeneities and the coexistence of two-phase state in these

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compositions. In this work, considering that the $Dy^{3+}$ ion has a larger magnetic moment ($\mu_{eff} \sim 10.6\mu_B$) and a smaller radius (1.07 Å) and the substitution of $Dy^{3+}$ ions at La sites will influence the structural and electrical and magnetic properties of the system, we investigate the magnetoresistance and magnetostriction effects of $(La_{1-x}Dy_x)_{2/3}Sr_{1/3}MnO_3$ (LDSMO) near $T_C$. The experimental results indicate that the LDSMO system exhibits not only a colossal magnetoresistance (CMR) but also a large VMS near $T_C$.

## 2. Experimental details

Polycrystalline samples of $(La_{1-x}Dy_x)_{2/3}Sr_{1/3}MnO_3$ ($0 \leq x \leq 0.6$) were prepared by solid-state reaction method. Appropriate amounts of high-purity $La_2O_3$, $Dy_2O_3$,$SrCO_3$ and $Mn_3O_4$ powders were weighted, ground carefully and then pressed into discs under 150 MPa and presintered at 1000 °C for 10 h in air. Then, they were crushed, ground again, pressed into pellets with 16 mm in diameter, and then sintered at 1280 °C for 10 h in air. The x-ray diffraction (XRD) analysis was carried out with a D-Max Rigaku system with the Cu K$\alpha$ radiation. The resistivities of the samples as a function of temperature were measured by the standard four-probe method with and without a magnetic field of 50 kOe using a Model-6000 type Physical Property Measurement System (PPMS). The MR is defined as $\Delta\rho/\rho = (\rho_H - \rho_0)/\rho_H$, where $\rho_H$ and $\rho_0$ represent the resistivities in the fields of $H$ and zero, respectively. The MS was measured by the strain gauge method. At the measurement one gauge was glued to the surface of the sample, the other gauge was glued to fused quartz, and both the gauges located in identical magnetic field and temperature. The longitudinal ($\lambda_{//}$) and transverse MS ($\lambda_\perp$) were measured parallel and perpendicular to the direction of the magnetic field, respectively. The volume ($\omega$) and anisotropic ($\lambda_t$) magnetostriction were calculated by formulas $\omega = \lambda_{//} + 2\lambda_\perp$ and $\lambda_t = \lambda_{//} - \lambda_\perp$. Magnetic properties were measured by a Quantum Design MPMS-XL superconducting quantum interference device (SQUID) magnetometer.

## 3. Results and discussion

Fig. 1 shows XRD patterns of the selected samples with $x = 0.1, 0.2, 0.3$ and 0.4. The sample with $x = 0.1$ exhibits a set of diffraction peaks of typical rhombohedral phase which accords with the earlier report [16]. With increasing $x$ the sample undergoes a structural transition from a rhombohedral phase to an orthorhombic phase which is also shown in the inset of Fig. 1. For $x = 0.3$ and 0.4 sample, the crystal structure exhibits a complete orthorhombic phase. From the inset of Fig. 1 it can be seen that the diffraction peak shifts to higher $2\theta$ angle, which indicates the decrease of the lattice constant with increasing $x$. It is attributed to the substitution of $Dy^{3+}$ with a smaller radius (1.07 Å) than that of $La^{3+}$ (1.22 Å), which results in the decrease of the average radius of A-site.

The $T_C$ values of the samples determined from the maximum point of the dM/dT in $M$–$T$ curves are 386, 337, 286, 229 and 177 K for $x = 0$, 0.1, 0.2, 0.3 and 0.4 samples, respectively. $T_C$ decreases monotonously which agrees with the report by Hwang et al. [17]. Fig. 2 shows the temperature dependence of the DC magnetization measured by a zero-field-cooling (ZFC) run and a field-cooling (FC) run at $H = 100$ Oe for $x = 0.3$. The inset of Fig. 2 shows the temperature dependences of reciprocal susceptibility ($1/\chi$) for $x = 0.3$, which exhibits a feature of antiferromagnetism [18]. The minimum of $1/\chi$ at 15 K observed for $x = 0.3$ should be attributed to an additional A-type antiferromagnetic coupling between $La^{3+}/Dy^{3+}$ and $Mn^{3+}/Mn^{4+}$ sublattices, as pointed out by Xu et al. [19]. Mitra et al. [20] stated that a spin-glass state also exists at low temperature in bulk Dy-doped LSMO samples as the case in Tb-doped LCMO samples. However, the argument on the initial susceptibility $\chi_{ac}$ versus temperature at different

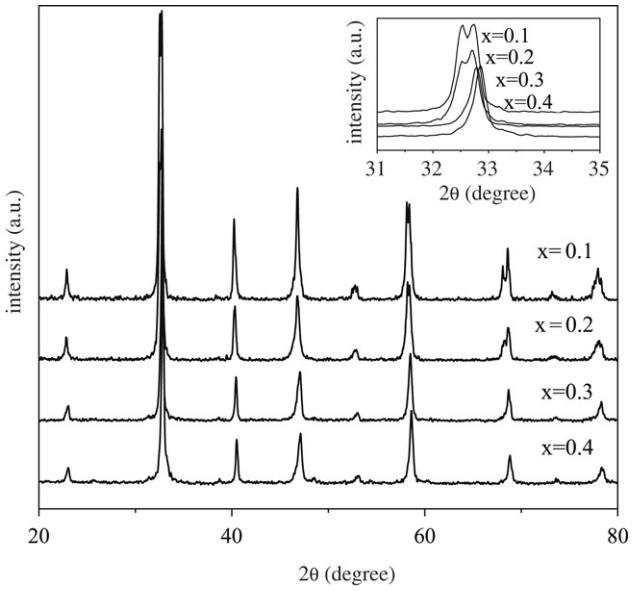


**Fig. 1.** XRD patterns of $x = 0.1, 0.2, 0.3$ and 0.4 samples. The inset shows the amplified patterns of the strongest peak.

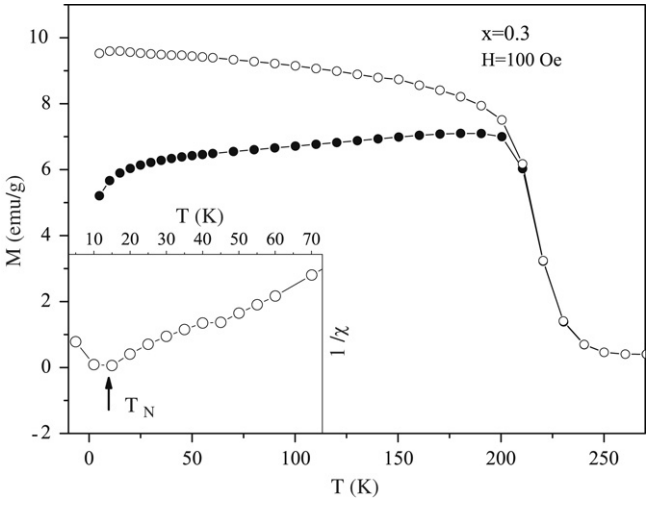


**Fig. 2.** ZFC (closed circle) and FC (open circle) magnetization curves at 100 Oe for $x = 0.3$ sample. The inset shows the temperature dependence of $1/\chi$.

frequencies (see Fig. 3 in Ref. [20]) is not so convincing, because one of typical features of a spin-glass state is that the maximum of $\chi_{ac}$ monotonously decreases with increasing frequency [21], but the maximum of $\chi_{ac}$ measured at 120 and 12 Hz by Mitra et al. were nearly the same. In addition, a larger tolerance factor $t$ also makes the existence of a spin-glass state in LDSMO unlikely. According to a universal phase diagram at low temperatures as a function of $t$ applicable to manganites including Dy-, Tb-, Pr-, and Y-doped LCMO (see Fig. 8 in Ref. [22]) given by Terai et al. [22], a spin-glass insulator state only exists in a range of $T < 50$ K and $t < 0.913$. For our LDSMO samples, the calculated $t$ value decreases from 0.944 for $x = 0$ to 0.922 for $x = 0.6$. These $t$ values are obvious greater than 0.913 and beyond the range required for the existence of a spin-glass insulator state in manganites. Fig. 3 shows $M$–$H$ curves at different temperatures. Magnetization increases with decreasing temperature and approaches to the saturation at 140 K and 10 kOe. As the temperature decreases further, $M$ at low magnetic field $H < 800$ Oe decreases slightly and $M$ at $H > 800$ Oe increases with $H$ and is not saturated up to 10 kOe. This implies that the magnetic moments in $Dy^{3+}$ and $Mn^{3+}/Mn^{4+}$ sublattices