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LETTER TO THE EDITOR

$Sm_{1-x}Sr_xMnO_3$ manganites: unusual magnetic, electric and elastic properties due to phase separation

A I Abramovich[1], **L I Koroleva and A V Michurin**

Physics Department, M V Lomonosov Moscow State University, Vorobyevy Gory, Moscow 119899, Russia

E-mail: abramovich@ofef343.phys.msu.su

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Abstract

Colossal magnetoresistance (CMR) ($\Delta\rho/\rho$) and giant negative volume magnetostriction (ω) have been observed in the Curie temperature region of $Sm_{1-x}Sr_xMnO_3$ manganites, for $x = 0.33$ compounds containing ferromagnetic (FM) and A-type antiferromagnetic (AFM) clusters and for $x = 0.4, 0.45$ compounds containing FM and both types of AFM clusters (A-type and charge-ordering (CO) type). For $x = 0.33$, the magnetization σ, $\Delta\rho/\rho$ and ω increase smoothly with magnetic field increase and saturation of $\Delta\rho/\rho$ and ω is not achieved. Isotherms of σ, $\Delta\rho/\rho$ and ω for $x = 0.4$ and 0.45 show another behaviour: sharp jumps of σ, $\Delta\rho/\rho$ and ω take place at $H_{C1} < H < H_{C2}$, and saturation is achieved at $H > H_{C2}$. We consider that the reason for CMR and giant magnetostriction being observed in the compounds investigated is the increase of the FM phase volume under the action of the magnetic field. For $x = 0.33$ this increase is smooth because it arises from FM phase 'sprouting' on FM layers of A-type AFM phase. For $x = 0.4$ and 0.45 the increase of the volume of the FM part arises from CO clusters with CE-type AFM structure too. In this case, CO clusters are completely transformed to the FM state with a large saturation magnetization σ_s which is equal to ~70% of σ_s at $T = 1.5$ K. This transition is accompanied by crystal structure reconstruction that is manifested in both the temperature and magnetic field dependences of the anisotropic magnetostriction.

1. Introduction

Manganites with colossal magnetoresistance (CMR) are attracting considerable attention because of their very rich physics and the prospects for their application in spin electronics. In this sense, $Sm_{1-x}Sr_xMnO_3$ system is especially interesting. It was found that the $x = 0.45$ compound shows not only CMR of ~42%, but also giant magnetostriction of $\sim 10^{-3}$ under

[1] Author to whom any correspondence should be addressed. Tel: +7(095) 9392847, Fax +7(095) 9328820.

a relatively low magnetic field of 8.4 kOe in the region of the Curie temperature T_C [1, 2]. Recent neutron diffraction and electron diffraction studies of the $Sm_{1-x}Sr_xMnO_3$ system [3, 4] have shown that a magnetic inhomogeneous state consisting of ferromagnetic (FM) clusters, A-type antiferromagnetic (AFM) clusters and charge-ordering (CO) AFM clusters is present for $x = 0.4, 0.45$. Both FM and A-type AFM clusters break down in the T_C-region and CO AFM clusters remain at $T > T_C$. The $x = 0.33$ compound contains FM and A-type AFM clusters and CO AFM clusters are absent from it. Thus, making a comparison of magnetic, electric, galvanomagnetic, elastic and magnetoelastic properties of $x = 0.33$ compounds with those of $x = 0.4, 0.45$ compounds will allow us to establish their peculiarities connected with the presence of CO clusters and clarify the question of the nature of their CMR and giant magnetostriction. With this aim, we have studied the magnetization, thermal expansion, magnetostriction, electrical resistivity and magnetoresistance of $Sm_{1-x}Sr_xMnO_3$ ($x = 0.33, 0.4, 0.45$) manganites.

2. Synthesis and experimental details

The ceramic samples were prepared by standard ceramic techniques. The phase composition and lattice parameters were controlled by means of x-ray diffraction. The ceramics were found to be pure single-phase perovskite with the orthorhombic *Pnma* structure. The magnetization measurements were performed with a SQUID magnetometer and the electrical resistance measurements by a four-probe method. The strain gauge technique was used for measurements of the magnetostriction and linear thermal expansion. Isotherms of the parallel ($\lambda_\parallel$) and perpendicular ($\lambda_\perp$) magnetostrictions with respect to the applied magnetic field (H) were measured. The anisotropic (λ_t) and volume (ω) magnetostrictions have been calculated as $\lambda_t = \lambda_\parallel - \lambda_\perp$ and $\omega = \lambda_\parallel + 2\lambda_\perp$. For $x = 0.33$ and 0.45 compounds the magnetostriction under a magnetic field up 15 T was measured in the laboratory of Professor M R Ibarra (Zaragosa, Spain).

3. Results and discussion

The T_C-values, determined from the AC initial magnetic susceptibility measurements in a magnetic field of 1 Oe amplitude and 8 kHz frequency, are 79, 112, 126 K for $x = 0.33, 0.4, 0.45$ respectively. It should be noted that here the notion of T_C is very relative. It is the temperature of the destruction of the FM ordering within the FM part of the sample. A sharp maximum in the temperature dependence of the resistivity (ρ) is found near T_C for all compounds. The MR, determined as $\Delta\rho/\rho = [\rho(H) - \rho(H = 0)]/\rho(H = 0)$, achieves colossal values of 83, 72 and 42% for $x = 0.33, 0.4, 0.45$ respectively in a magnetic field of 8.4 kOe. A sharp change in the linear thermal expansion is observed near T_C for $x = 0.4, 0.45$ with a corresponding volume change $\Delta V/V \sim 0.1\%$. For $x = 0.33$ the change in linear thermal expansion in the T_C-region happens more smoothly. Figure 1 displays the temperature dependence of both the magnetization and the volume magnetostriction at selected magnetic fields for $x = 0.45$ ((a), (b)) and for $x = 0.33$ ((c), (d)). For the $x = 0.40$ compound the behaviours of both the temperature and the magnetic field dependences of σ, ω and $\Delta\rho/\rho$ are similar to those for the $x = 0.45$ compound. Giant negative volume magnetostriction has been observed for all compounds: at $T \leqslant T_C$ for $x = 0.33$ compounds having A-type AFM clusters only; and at $T \geqslant T_C$ for $x = 0.4$ and 0.45 compounds having both types of AFM cluster. For all compounds, the $\omega(T)$ curves pass through a minimum in the T_C-region. For $x = 0.33$, σ, $\Delta\rho/\rho$ and ω increase smoothly with magnetic field increase and saturation of $\Delta\rho/\rho$ and ω is not achieved under magnetic fields up to 130 kOe (figure 2). Isotherms of σ, $\Delta\rho/\rho$ and ω

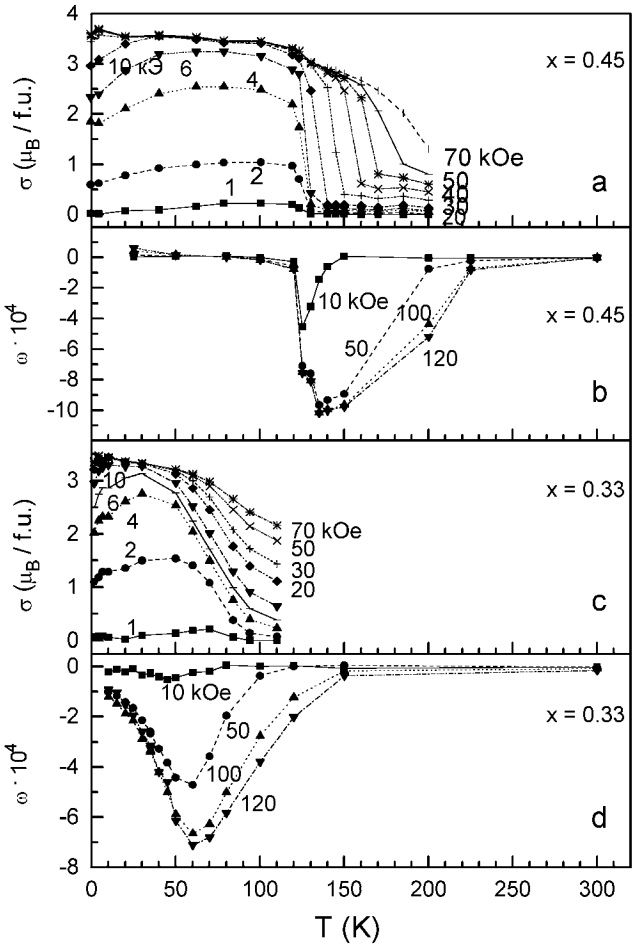

Figure 1. Temperature dependences of the magnetization and volume magnetostriction, at selected magnetic fields, for $x = 0.45$ ((a), (b)) and for $x = 0.33$ ((c), (d)) compounds.

for $x = 0.4$ and 0.45 show another behaviour (figure 3). (The isotherms of $\Delta\rho/\rho$ in figures 2 and 3 have not been shown.) The sharp jumps of σ, $\Delta\rho/\rho$ and ω take place within the critical magnetic field region $H_{C1} < H < H_{C2}$, and they reach saturation at $H > H_{C2}$. One notes that both H_{C1} and H_{C2} defined using the magnetization measurements coincide with the ones defined using magnetostriction and magnetoresistance measurements at the same temperature (figure 4). Also, metastability of σ, ρ and ω occurs at $H_{C1} < H < H_{C2}$.

The observed properties are explained by the existence of the phase separation due to the strong s–d exchange [5, 6]. In this case, the crystal is separated into AFM and FM parts and charge carriers are located in the latter. Yanase and Kasuya [7] have shown that the lattice parameters are reduced inside the FM part of crystal. The increase in the volume of the FM part under the action of a magnetic field leads to CMR and giant magnetostriction for $x = 0.40$ and 0.45 compounds like for $x = 0.33$ compounds. For $x = 0.33$ this increase is smooth because it arises from FM phase 'sprouting' on FM layers of A-type AFM phase. For $x = 0.4$ and 0.45 the increase of the volume of the FM part occurs due to CO clusters with CE-type AFM structure too, in which spins of neighbouring Mn ions are AFM ordered. For this reason the transition of CO clusters to the FM state must occur at magnetic field energy different

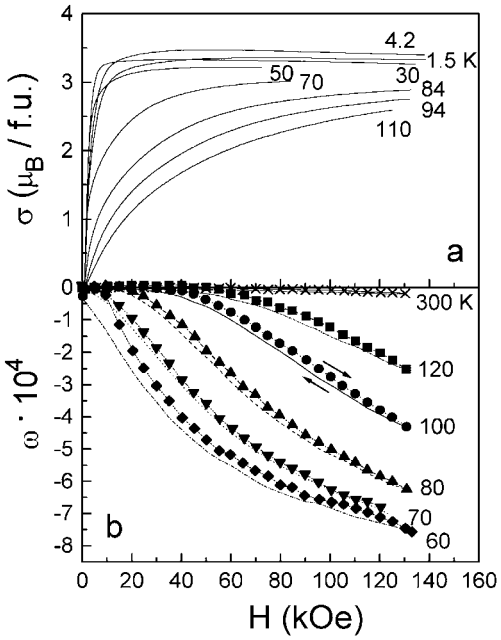

Figure 2. Magnetization (a) and magnetostriction (b) isotherms at selected temperatures for the $x = 0.33$ compound.

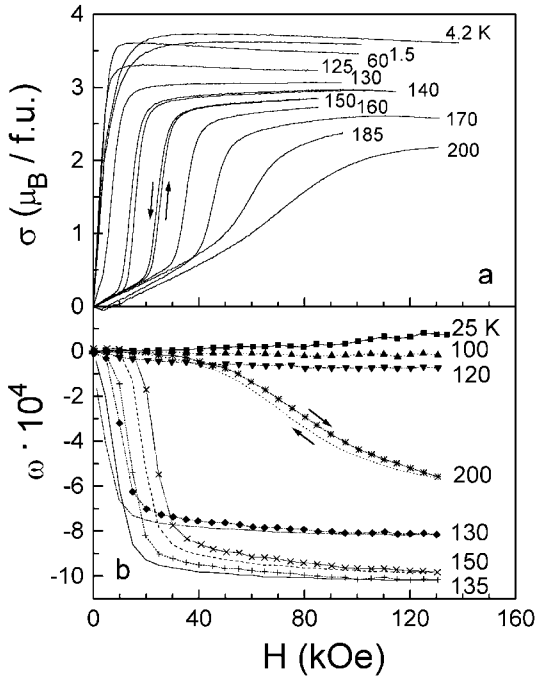

Figure 3. Magnetization (a) and magnetostriction (b) isotherms at selected temperatures for the $x = 0.45$ compound.

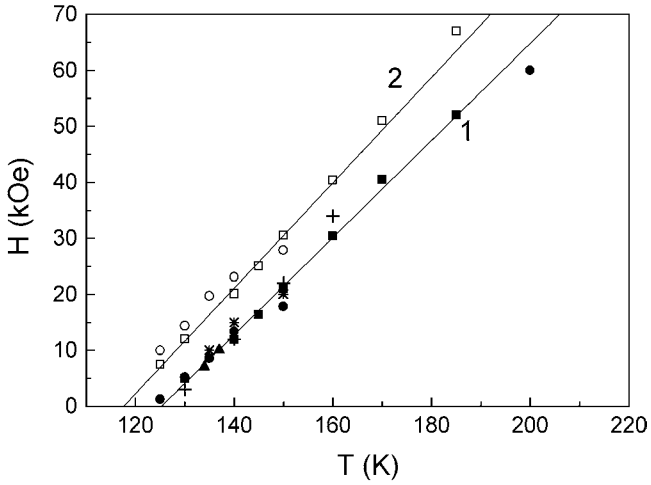

Figure 4. Temperature dependences of critical magnetic fields H_{C1} (1) and H_{C2} (2) for the $x = 0.45$ compound. The critical fields were obtained from magnetization measurements (■, □), magnetostriction measurements (●, ○), magnetoresistance measurements (▲), anisotropic magnetostriction measurements (∗) and the magnetoresistance measurements of [8] (+; H_{C1} only).

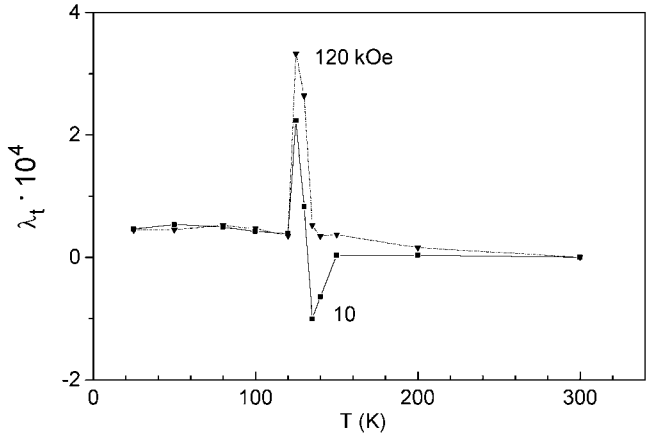

Figure 5. Temperature dependences of the anisotropic magnetostriction at selected magnetic fields for the $x = 0.45$ compound.

from zero. The sharp jumps in the $\sigma(H)$, $\omega(H)$, $\Delta\rho/\rho(H)$ curves and their subsequent saturation indicate that the transition of CO clusters to the FM phase has a threshold character. In this case all CO clusters are completely transformed to the FM state with a large saturation magnetization σ_s which is equal to ∼70% of σ_s at $T = 1.5$ K (see figure 3). The transition of AFM CO clusters to the FM state under the action of a magnetic field must be accompanied by crystal structure reconstruction for $x = 0.4$ and 0.45 compounds that must be manifested in the anisotropic magnetostriction behaviour. Indeed, we have observed peculiarities in both the temperature and magnetic field dependences of the anisotropic magnetostriction, which are presented in figures 5 and 6 for $x = 0.45$ compounds. One can see that λ_t is small throughout the whole temperature interval; it increases in the T_C-region and pass through a maximum, then changes sign at T_C and passes through a minimum near the temperature of the CO phase destruction (T_{CO}). At $T \geqslant T_C$, $\lambda_t(H)$ isotherms pass through a minimum at some magnetic

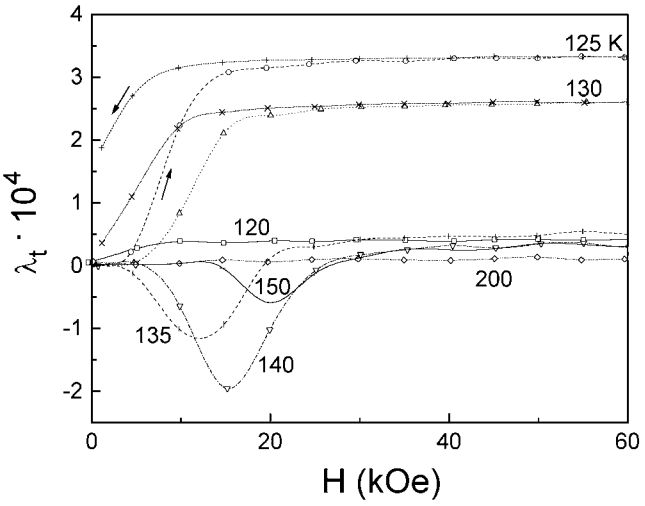

Figure 6. Magnetic field dependences of the anisotropic magnetostriction at selected temperatures for the $x = 0.45$ compound.

field which coincides with H_{C1} defined on the basis of magnetization, magnetostriction and magnetoresistance measurements (see figure 4). At $H > H_{C2}$, when the FM state of the sample is achieved, λ_t is positive throughout the whole temperature interval and passes through a maximum near T_C (see figure 5).

In summary, we have shown that the CMR and giant magnetostriction observed in $Sm_{1-x}Sr_xMnO_3$ manganites have the same nature and are caused by increase of the FM phase volume under the action of a magnetic field. For $x = 0.33$ this increase is smooth because it arises from FM phase 'sprouting' on FM layers of A-type AFM phase. For $x = 0.4$ and 0.45 the increase of the volume of the FM part arises from CO clusters with CE-type AFM structure too. In this case CO clusters are completely transformed to the FM state, whose saturation magnetization σ_s is equal to $\sim$70% of σ_s at $T = 1.5$ K. This transition is accompanied by crystal structure reconstruction that is manifested in both the temperature and magnetic field dependences of the anisotropic magnetostriction.

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