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Physics Letters A 346 (2005) 321–326
PHYSICS LETTERS A
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Internal friction evidence of uncorrelated magnetic clusters in electron-doped manganite $Sr_{0.8}Ce_{0.2}MnO_3$

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Received 8 June 2005; accepted 29 July 2005

Available online 15 August 2005

Communicated by J. Flouquet

Abstract

Electron-doped manganite $Sr_{0.8}Ce_{0.2}MnO_3$ has been systematically investigated by X-ray diffraction, electronic transport, magnetic, internal friction, and Young's modulus experiments. The X-ray diffraction result indicates that the compound remains tetragonal (I4/mcm) structure at room temperature. Due to the strong Jahn–Teller (JT) distortion, the ground state is antiferromagnetic (AFM) insulator. Below 20 K, a spin-glass (SG) state dominates at low temperatures. In the paramagnetic (PM) region, an internal friction peak at around 250 K, which is characteristic of relaxation, has been observed. Under applied magnetic field, the internal friction peak moves to higher temperature, which is suggested to originate from the formation of ferromagnetic (FM) clusters in PM region. In addition, the softening of Young's modulus in the vicinity of AFM transition temperature is interpreted in terms of the strong electron–phonon interaction.

PACS: 75.47.Lx; 75.30.Kz; 62.40.+i

Keywords: Manganites; Magnetic clusters; Phase separation; Internal friction

1. Introduction

Hole-doped manganese oxides $R_{1-x}A_xMnO_3$ (R = rare-earth ions; A = divalent ions such as Ca, Sr, Ba, Pb, etc.) with a perovskite structure have stimulated considerable scientific and technological interest because of their exotic electronic and magnetic properties [1–4]. These manganites present a colossal magnetoresistance (CMR) effect and the characteristic feature of the CMR manganites is the strong interaction between charge carriers in the e_g band, localized spins of t_{2g} electrons, and the crystal lattice. Several theoretical models such as the double-exchange (DE) mechanism [5], small polarons, electron-lattice coupling, and especially, the Jahn–Teller (JT) type interaction [6–8], were proposed to explain the physics of CMR properties.

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0375-9601/$ – see front matter
doi:10.1016/j.physleta.2005.07.084

In contrast to the hole-doped materials, there have been a few electron-doped materials studied in this system. Zeng et al. [9] reported large negative magnetoresistance in electron-doped $Ca_{1-x}Ce_xMnO_3$ system. Low Ce doping stabilizes a ferromagnetic (FM) component in addition to a persistent antiferromagnetic (AFM) component. Higher Ce doping ($x \geqslant 0.075$) induces a charge ordering (CO) transition. The physical properties of $Ca_{1-x}Ce_xMnO_3$ are similar to those of hole-doped manganites [9,10]. However, Sundaresan et al. [11] have reported that the electron-doped $Sr_{1-x}Ce_xMnO_3$ does not show large negative magnetoresistance. Also the magnetic susceptibility (χ) versus T curve for $x > 0.10$ samples shows a broad maximum, which is absent in hole-doped manganites. They have explained this unusual behavior of χ using the dilute AFM model. Recently, Mandal et al. [12] prepared Ce doped $Sr_{1-x}Ce_xMnO_3$ by using a two step method and reported that the system did not show any long-range magnetic ordering but spin-glass like behavior at low temperatures for $x \geqslant 0.10$. For $x = 0.25$ and 0.35 samples, it shows large negative magnetoresistance over a wide range of T. The differences of the results in the last two papers should be due to the use of different ways for the sample preparation.

Anyway, the different electronic, and magnetic properties between electron-doped $Sr_{1-x}Ce_xMnO_3$ and hole-doped manganites have not fully been elucidated so far. Therefore, a detailed study of the crystallographic and magnetic phase diagram is needed. On the other hand, a potential intrinsic magnetic inhomogeneities in $Sr_{1-x}Ce_xMnO_3$ system should arouse our attention, since the evidence of inhomogeneities in manganites is very strong both in theory and experiments [13,14]. In this Letter, we studied the microscopically and intrinsically inhomogeneous magnetic phase in $Sr_{0.8}Ce_{0.2}MnO_3$ sample, for which a very broad maximum in $\chi(T)$ curve appears implying the dilute antiferromagnetism [11]. Here we use internal friction measurement technique to study the microscopic mechanism of magnetic phase transitions. The measurements, as discussed below, enable to obtain important information about magnetic phase separation.

The internal friction measuring technique is a nondestructive but very sensitive tool in studying defects and microscopic processes in solids including electron strongly correlated materials, such as cuprate high temperature superconductors and manganites [15–20]. By means of internal friction measurements, one may determine whether the microscopic process is phase transition or relaxation, and some other quantitative information such as activation energy and the relaxation time at infinite temperature can be obtained. In addition, the concomitant Young's modulus reflects the information of the lattice variation. The strong coupling of electron–phonon can be studied by the Young's modulus measurements. For $Sr_{0.8}Ce_{0.2}MnO_3$ sample, a remarkable peak in the internal friction curve is observed in the paramagnetic (PM) region, which is attributed to the formation of magnetic clusters. Furthermore, this peak is characteristic of thermally activated relaxation. The anomalous Young's modulus properties imply the electron–phonon interaction due to the JT effect may play an important role in the sample.

2. Experimental details

Polycrystalline $Sr_{0.8}Ce_{0.2}MnO_3$ sample was synthesized by the standard solid-state reaction. Stoichiometric precursor powders $SrCO_3$, CeO_2, MnO_2 and Cr_2O_3 were mixed and ground, then fired for 24 h at 1050 °C. The resultant powder was then pressed into small pellets and sintered at 1200 °C for 24 h and then at 1350 °C for another 24 h. After the final grinding, the powder was pressed into bars with the dimension of $65.0 \times 4.5 \times 1.0$ mm^3 (for internal friction and Young's modulus measurements), sintered at 1400 °C for 24 h and slow-cooled to room temperature.

The room temperature X-ray diffraction (XRD) measurement was taken in Philips X'pert PRO X-ray diffractometer with Cu K_α radiation. The structural parameters were obtained by fitting the experimental data of XRD using the standard Rietveld technique. The magnetic measurements were carried out with a Quantum Design superconducting quantum interference device (SQUID) MPMS system. The resistivity ρ was measured by means of a Quantum Design physical properties measurements system (PPMS) using the standard four-probe method. Internal friction $Q^{-1}(T)$ and Young's modulus $E(T)$ were measured by the free decay method of a resonant bar in acoustic frequency range with magnitude of kHz using warming mode in a vacuum environment at the rate of 1 K/min. The in-