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Anomalous sound velocity behavior of $La_{1-X}Ca_XMnO_3$ ($X\sim0.48$) in applied field

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Abstract

The sound velocity $v_s(T)$ has been investigated for $La_{1-X}Ca_XMnO_3$ ($0.46\leqslant X\leqslant0.50$) in applied fields up to $\mu_0H = 5\,T$. The characteristic $v_s(T)$ softening and the subsequent hardening are observed below the ferromagnetic transition temperature T_c. The gradual $v_s(T)$ softening above the charge-order (CO) transition temperature T_{CO} and the $v_s(T)$ hardening just below T_{CO} are also observable. The $v_s(T)$ softening above T_{CO} is at first enhanced and then reduced with increasing applied field. These results can be understood on the basis of the enhanced spin–lattice and charge–lattice interaction.

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Keywords: $La_{1-X}Ca_XMnO_3$; Sound velocity; Spin–lattice interaction; Electron–lattice interaction

1. Introduction

Carrier-doped perovskite manganites exhibit a variety of dramatic phenomena caused by the competition or cooperation of the charge order (CO), orbital order (OO), Jahn–Teller (JT) effect, double exchange (DE) and superexchange (SE) interaction, etc. Important information on the electron–lattice interaction related to the orbital and charge degrees of freedom can be obtained from the sound velocity $v_s(T)$ measurement. For $La_{1-X}Sr_XMnO_3$ (LSMO) and $La_{1-X}Ca_XMnO_3$ (LCMO) systems, we reported the $v_s(T)$ anomalies associated with CO under the zero magnetic field [1–3]. In LCMO, there is a phase boundary around $X = 0.50$ between the ferromagnetic (FM) metal and the CO-antiferromagnetic (CO-AFM) insulator, and the volume fraction of each phase in actual specimens changes depending on the Ca concentration X, the applied field μ_0H and the temperature T. In this paper, we report the anomalous $v_s(T)$ for LCMO system ($X\sim0.48$) in applied fields and discuss the lattice dynamics coupled with spins and charge carriers.

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2. Experimental

$La_{1-X}Ca_XMnO_3$ ($0.46\leqslant X\leqslant0.50$) samples were fabricated by a conventional solid-state reaction method [2]. $v_s(T)$ was measured by a pulse-superposition method using $\sim$7 MHz longitudinal waves in applied fields up to $\mu_0H = 5\,T$ in the processes of field cooling (FC) and field warming (FW) from 4.2 to 300 K. The magnetization $M(T)$ was measured by a SQUID magnetometer.

3. Results and discussion

Fig. 1 shows $v_s(T)$ in zero field and $M(T)$ in 0.5 T for the $X = 0.46$ and 0.47 samples. For $X = 0.46$, the FM transition occurs at $T_c = 235\,K$ and the $v_s(T)$ upturn was observed below T_c with a faint initial softening. The $v_s(T)$ upturn indicates the strong coupling of the lattice to the spins. The $X = 0.47$ sample shows two-step phase transitions; T_c decreases to 230 K and the CO-AFM phase appears at lower temperatures. The $v_s(T)$ softening just below T_c increases, compared with that for $X = 0.46$, which may suggest the enhancement of the spin fluctuation. The $v_s(T)$ hardening with a large hysteresis takes place at

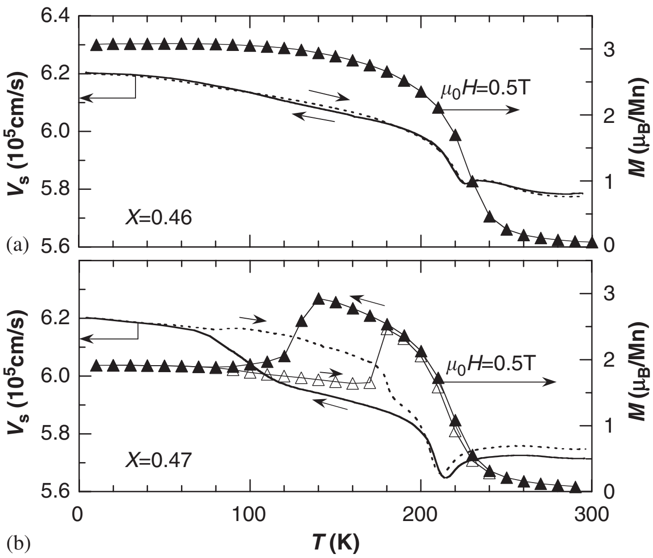

Fig. 1. $v_s(T)$ in zero field and $M(T)$ in 0.5 T for (a) the $X = 0.46$ and (b) $X = 0.47$ samples.

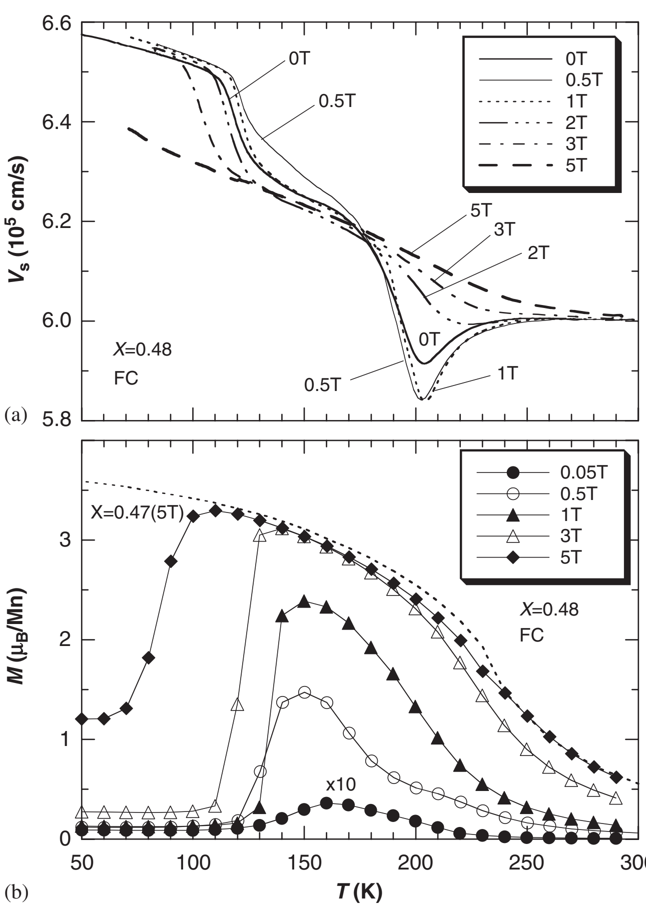

Fig. 2. (a) $v_s(T)$ and (b) $M(T)$ for the $X = 0.48$ sample in the field-cooling (FC) runs.

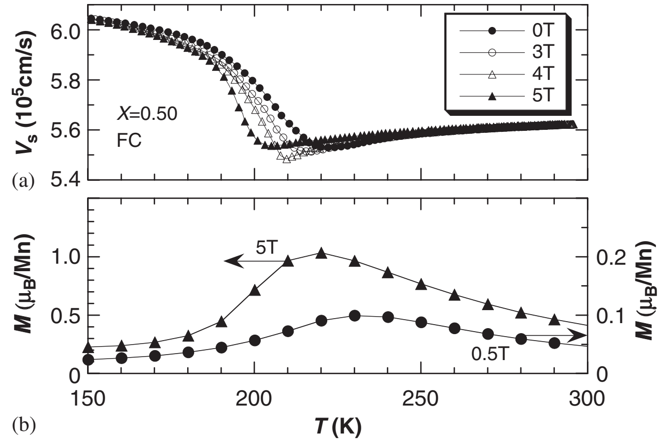

Fig. 3. (a) $v_s(T)$ of the $X = 0.50$ sample in the (FC) runs and (b) the $M(T)$ in $\mu_0 H = 0.5$ and 5 T.

$T_{CO} = 120$ K in FC and at 180 K in FW run, where the FM to CO-AFM phase transition occurs. The approach of X to $\frac{1}{2}$ with resultant Mn^{3+}:Mn^{4+}~1:1, makes the CO-AFM state stable. The second hardening in $v_s(T)$ at the low temperatures is caused by the appearance of the CO-AFM state.

Fig. 2 shows (a) $v_s(T)$ and (b) $M(T)$ for the $X = 0.48$ sample in the FC runs. The v_s softening just below $T_c = 225$ K for $\mu_0 H = 0$ T promptly becomes deep with increasing applied field up to 1 T, suggesting the enhancement of the spin fluctuation. This softening is reduced suddenly for $\mu_0 H \geqslant 2$ T and is wiped out for $\mu_0 H = 5$ T due to the nearly complete alignment of the FM moment. The $v_s(T)$ upturn around 100 K due to the CO-AFM transition shifts to a lower temperature with increasing applied field.

Fig. 3(a) shows $v_s(T)$ for the $X = 0.50$ sample in the FC runs. Fig. 3(b) shows $M(T)$ for $\mu_0 H = 0.5$ and 5 T, showing the decrease of T_{CO} (defined by the $M(T)$ maximum) with increasing magnetic field. The gradual $v_s(T)$ softening above T_{CO} coming from the charge fluctuation between Mn^{3+} and Mn^{4+} ions is enhanced up to 4 T and suddenly reduced in the field of 5 T. These results suggest that the CO-AFM phase at $X = 0.50$ is stable even under the strong magnetic field.

In summary, the FM and CO-AFM transitions of $La_{1-X}Ca_XMnO_3$ samples (X~0.48) were monitored by the sound velocity $v_s(T)$ in magnetic fields. With increasing applied field, the spin and/or charge fluctuations around each phase boundary are at first enhanced and then reduced when the FM or CO-AFM state is stabilized.

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