Magnetic and submillimeter spectroscopy study of phase transitions in $La_{1-x}Sr_xMnO_3$ perovskites: T–x phase diagram

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Lanthanum manganites doped by the divalent ions Sr (Ca) exhibit various interesting phenomena such as a colossal negative magnetoresistance, magnetic, structural and metal-insulator phase transitions, a charge or polaron ordering [1–3]. The parent compound $LaMnO_3$ is a Mott insulator and has a canted antiferromagnetic layer structure. The substitution of La^{3+} ions by Sr^{2+} ions results in a transition from antiferromagnetic insulating state to a ferromagnetic metallic state at $x \geqslant 0.17$, which is stabilized by a double exchange. The crystal structure in this case is also changed from the orthorhombic to rhombohedral symmetry [1]. Complicated phase transformations are expected in the intermediate concentrations $0.1 \leqslant x \leqslant 0.15$ [2,3]. In this work we performed complex investigations of various magnetic and structural transitions in $La_{1-x}Sr_xMnO_3$ manganites by means magnetic and electric static and submillimeter dynamic measurements.

Single crystals of $La_{1-x}Sr_xMnO_3$ ($0 \leqslant x \leqslant 0.45$) were grown by the floating zone method with radiation heating. The transmission and phase shift spectra of thin plane-parallel plates were measured using a quasi-optical submillimeter backward-wave-oscillator technique [4] in the frequency range $\nu = 3$–$33\ cm^{-1}$, that allowed to obtain a dynamic dielectric permittivity ε' and conductivity σ'. Resistance $\rho(T)$ were measured using the four-probe method at temperatures 4.2–1050 K. Magnetization $M(T,H)$ and AC magnetic susceptibility $\chi_{AC}(T)$ measurements were performed at $T = 4.2$–400 K in magnetic field up to 12 kOe.

Temperature dependence of the resistance $\rho(T)$ are shown in the Fig. 1. The curves exhibit several kinds of anomalies (some of them were also observed in Ref. [1] at $T < 500$ K) assigned by different symbols and identified with the following transitions. (1) Ferromagnetic phase transition at T_C accompanied by a metal-semiconductor transition ($x \geqslant 0.1$), which assigned by arrows. This transition also manifests itself distinctly as a sharp increase $\chi_{AC}(T)$ at T_C (Fig. 2c). (2) Structural phase transitions at T'_s between low-temperature orthorhombic (Jahn–Teller) O′ phase and high-temperature orthorhombic (pseudocubic) O* phase at $0 \leqslant x \leqslant 0.125$ (filled down triangular). The most strong anomaly in the $\rho(T)$ is observed for the pure $LaMnO_3$ at $T'_s \approx 750$ K and

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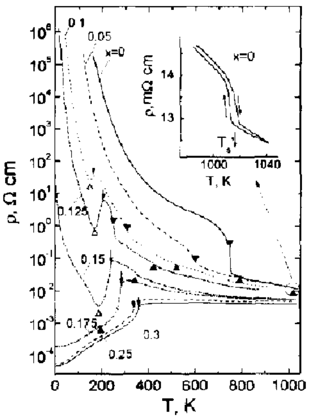

Fig. 1. Temperature dependence of the resistance in $La_{1-x}Sr_xMnO_3$. Inset: $\rho(T)$ behavior near a new structural transition in $LaMnO_3$. Symbols indicate various phase transitions (see text).

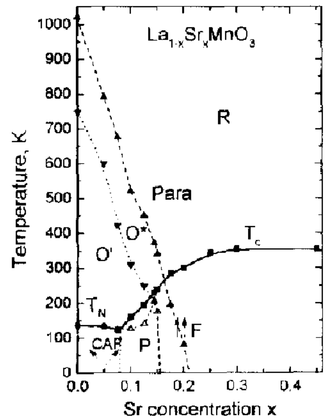

Fig. 3. Phase T–x diagram of $La_{1-x}Sr_xMnO_3$: (F) ferromagnetic phase, (CAF) canted antiferromagnetic phase, (O') Jahn–Teller orthorhombic phase, (O*) orthorhombic (pseudocubic) phase, (P) polaron ordered state.

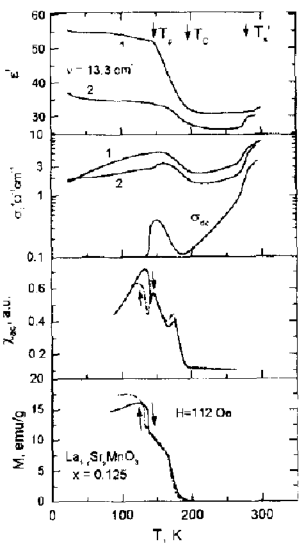

Fig. 2. Temperature dependence of the dielectric permittivity $\varepsilon'(13.3\ cm^{-1})$ (a), dynamic ($\sigma'(13.3\ cm^{-1})$) and static (σ_{DC}) conductivity (b), AC magnetic susceptibility (c) and magnetization (d) in $La_{0.875}Sr_{0.125}MnO_3$. Indices 1,2 for the ε' and σ' correspond to different radiation polarizations.

corresponds to the known Jahn–Teller transition accompanied by an orbital ordering and significant lattice distortions (see also Ref. [5]). In order to identified this transitions we used results of a neutron diffraction study [3] for $x = 0.125$. (3) Structural orthorhombic (O*) to rhombohedral (R) phase transitions at T_s and $0 \leqslant x \leqslant 0.22$ (filled up triangular). The $\rho(T)$ anomaly at T_s is too weak in this case and can be seen only for a suitable scale (inset in Fig. 1). (4) Transitions to a polaron ordered state (P) at T_p and $0.1 \leqslant x \leqslant 0.15$, determined by freezing of holes on the lattice sites (open up triangular). Such transitions were observed by neutron scattering in $La_{1-x}Sr_xMnO_3$ for $x = 0.1$ and 0.15 [2].

The results of the submillimeter measurements of the dielectric permittivity $\varepsilon'(T)$ and conductivity $\sigma'(T)$ at the frequency 13.3 cm^{-1} for $x = 0.125$, combined with static conductivity $\sigma_{DC}(T)$ AC magnetic susceptibility $\chi_{AC}(T)$ and magnetization $M(T)$ measurements, are displayed in Fig. 2. Curves 1,2 for the ε' and σ' correspond to the radiation polarization for a minimum and maximum transmission, respectively, that occurs due to a noticeable anisotropy of a crystal lattice. Observed features in the $\varepsilon'(T)$ and $\sigma'(T)$ at T_s', T_c and T_p, indicated by arrows, are in a reasonable agreement with corresponding static data. A significant increase of the $\varepsilon'(T)$ in the polaron ordered state (Fig. 2a) indicates on a noticeable transformation of a crystal lattice and, probably, a change of electron structure, which was observed recently by optical measurements [6]. We note also that σ' remains large enough at low temperatures in spite of localization of charge carriers which results in a significant lowering of the σ_{DC} (Fig. 2b). A similar behavior of the $\varepsilon'(T)$ and $\sigma'(T)$ was also observed for $x = 0.1$ and 0.15 (see also Ref. [7]).

The observed anomalies in ρ, χ_{AC}, M, ε' and σ' at various phase transitions are displayed in the form of the T–x phase diagram in Fig. 3, where solid lines correspond to magnetic transitions and doted and dashed lines-to the structural ones. The polaron ordered phase P is located approximately between $x = 0.08$ and 0.15. The phase diagram presents a general picture of the phase transitions in $La_{1-x}Sr_xMnO_3$ and in the whole, it agrees with corresponding data of Ref. [2,3].

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