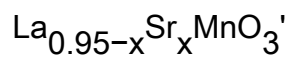


Reply to the comment on 'Low-temperature transport properties of non-stoichiometric



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REPLY

Reply to the comment on ‘Low-temperature transport properties of non-stoichiometric $\text{La}_{0.95-x}\text{Sr}_x\text{MnO}_3$ ’

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Received 22 April 2002

Published 5 September 2002

Online at stacks.iop.org/JPhysCM/14/8759

Abstract

The authors of the comment dispute that the shallow minimum, observed at low temperatures in different manganates, has the origin to the 3D enhanced electron–electron interaction. In this reply various arguments are used to show that the cause of this phenomenon is more probably the electron–electron interaction.

Rare-earth manganese perovskites show a rich variety of structural, magnetic and transport properties. One of the most interesting physical phenomena exhibited by these materials is a resistivity minimum observed at $T = T_{min} < 30$ K in doped perovskites [1–7] showing metallic conductivity. Most authors [1, 2, 6, 7] have found that the resistivity at $T < T_{min}$ follows a $\sim -T^{1/2}$ -dependence. The same temperature dependence can also be deduced from literature data, for example that of Roy *et al* [5] (figure 1). In order to explain the origin of the observed resistivity minimum the experimental data have been analysed taking into account:

- (a) the enhanced electron–electron (e–e) interaction in bulk weak disordered metallic systems [1, 2, 6, 7]; and
- (b) intergranular tunnelling of the polarized charge carriers between antiferromagnetically coupled neighbouring grains in polycrystalline ferromagnetic specimens [3, 4].

In our earlier paper [6] the observed minimum of the electrical resistivity in investigations of a single-phase bulk polycrystalline specimen of $\text{La}_{0.95-x}\text{Sr}_x\text{MnO}_3$ ($x = 0.10, 0.20, 0.30$) has been attributed to enhanced e–e interaction. It must be accepted, of course, that the reported results are only a limited subset of the data obtained in the systematic investigation of the system of $\text{La}_{y-x}\text{Sr}_x\text{MnO}_3$ ($y = 1.00, 0.95, 0.00 \leq x \leq 0.40$).

In order to provide a complete argument in support of the interpretation of the data presented in our earlier paper [6], it is necessary to report briefly the properties of the $\text{La}_{y-x}\text{Sr}_x\text{MnO}_3$ ($y = 1.00, 0.95; 0.00 \leq x \leq 0.40$) polycrystalline specimen investigated systematically, but not reported on fully in [6].

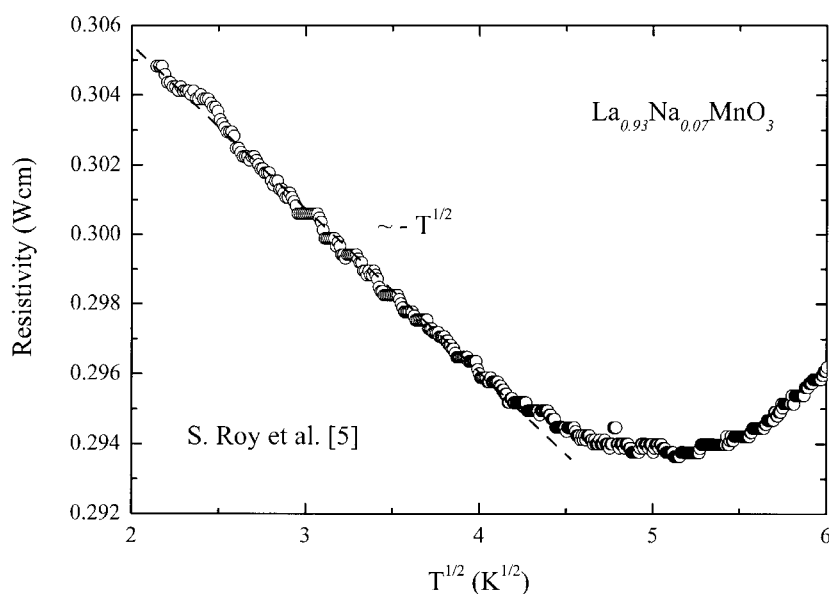


Figure 1. The resistivity data of the measurements made by Roy *et al* [5] for $\text{La}_{0.93}\text{Na}_{0.07}\text{MnO}_3$ compound as a function of $T^{1/2}$.

- (1) The perovskite powders were prepared using nitrate salts, with minimum purity of 99.9%, by a wet-chemical technique [8]. In addition to the phase composition identification and lattice parameter determination by means of x-ray diffraction, the $\text{La}_{0.95-x}\text{Sr}_x\text{MnO}_3$ cationic ratios derived were verified by ICP-AES within experimental uncertainty.
- (2) Metallographic and SEM examinations, figure 2, revealed well-densified specimens. Reduced porosity and a homogeneous grain structure can be seen, with a mean grain size decreasing from about $19 \mu\text{m}$ for $x = 0.10$ to $6 \mu\text{m}$ for $x = 0.30$. Thus the grain boundary area crossed by the polarized charge carriers should be greater for higher x . Therefore, tunnelling contributions, which have been found to be insensitive to composition [3, 4], could plausibly be expected to become stronger as x increases, leading to behaviour opposite to that observed for the depth of the resistivity minimum, as discussed below.
- (3) The resistivity of the specimens decreases significantly with increasing x . For $x = 0.20$ and 0.30 , it approaches that of corresponding single-crystalline compounds [9] within a factor of less than ten in the metallic regime. However, it is lower by two orders of magnitude at $T \leq T_c$ than the resistivity of polycrystalline $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$. Thus, the microstructural contributions to the resistivity should be of minor importance for the $\text{La}_{0.95-x}\text{Sr}_x\text{MnO}_3$ specimen investigated. This is apparently not the case for the $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ ceramics which have been used to test the applicability of the grain boundary model so far [3, 4]. $\text{La}_{0.85}\text{Sr}_{0.1}\text{MnO}_3$ shows metallic resistivity below 135 K, probably due to the vacant La sites, which might cause self-doping of this compound with additional Mn^{4+} ions.
- (4) The ZFC magnetic moment of $\text{La}_{0.95-x}\text{Sr}_x\text{MnO}_3$ at $H = 5 \text{ mT}$, obtained by a SQUID magnetometer, shows ferromagnetic behaviour with Curie temperatures $T_c = 200, 325$ and $>360 \text{ K}$ for the specimens with $x = 0.10, 0.20, 0.30$, respectively. The spontaneous magnetic moment shows a smooth decrease below T_c only for the $x = 0.10$ compound. No additional change can be recognized at $T < T_{min}$ for any of the specimens. It was

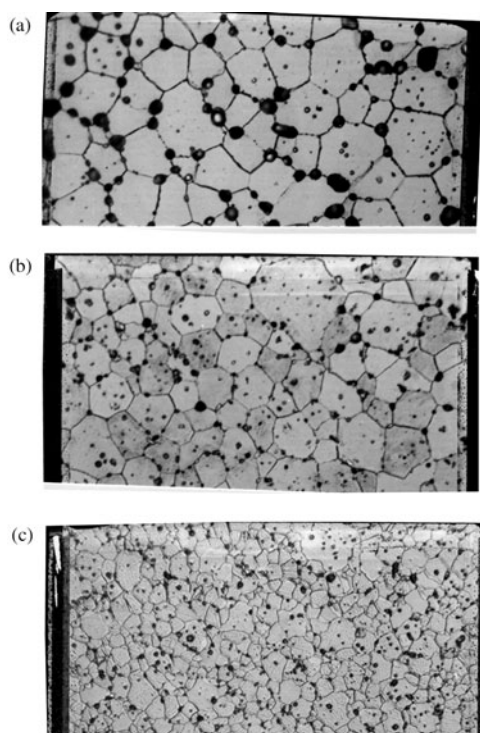


Figure 2. SEM micrographs of the compounds investigated: (a) $\text{La}_{0.85}\text{Sr}_{0.10}\text{MnO}_3$ (b) $\text{La}_{0.75}\text{Sr}_{0.20}\text{MnO}_3$ and (c) $\text{La}_{0.65}\text{Sr}_{0.30}\text{MnO}_3$.

- observed that the metal-insulator transition temperature (T_{IM}) is approximately equal to T_c for the specimens with $x = 0.20$ and 0.30 and $T_{max} = 135$ K for $x = 0.10$ respectively.
- (5) The electrical resistivity of the stoichiometric $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ ($x = 0.16, 0.20, 0.25, 0.35, 0.40$) polycrystalline specimens, prepared as above, exhibited metallic resistivity with a minimum at $T < 40$ K. The corresponding T_{min} varies approximately as $x^{-2/5}$ with Sr content x [10].

However, it is well known that the minimum and the $\sim -T^{1/2}$ -dependence on the resistivity are observed in weakly disordered bulk metallic systems [11] and amorphous alloys [12]. Such behaviour is attributed to the interference of e-e scattering with scattering from static inhomogeneities [13] in three dimensions.

Very recently Kumar *et al* [7], by measuring the resistivity $\rho(T, H)$ of single-phase 1000 Å thick films of $\text{La}_{0.70}\text{Ca}_{0.30}\text{MnO}_3$ as a function of temperature and magnetic field, found that:

- (a) At low temperatures the resistivity shows a $\sim -T^{1/2}$ -dependence with a minimum which shifts to higher temperatures as the magnetic field H increases, goes through a very broad maximum and at $H > 2$ T begins to decrease slightly (see figure 3 in [7]). It seems that the minimum persists and is affected only very weakly by the magnetic field.
- (b) The depth of the minimum increases with the magnetic field and above 3 T shows a tendency to saturate.
- (c) T_{min} , like the depths of the minima, is dependent on the resistivity of the films. The films with higher electrical resistivities show higher values of T_{min} and greater depths.

- (d) The coefficient of the $T^{1/2}$ -dependence at 5 T is larger than that in zero field (see figure 2 in [7]).

Thus, considering our results and those of Kumar *et al* [7], we can now discuss the objections of Rozenberg and Auslender regarding the origin of the minimum of the resistivity in the polycrystalline manganites.

- (i) Most investigations have found that the resistivity of manganites in the metallic state and at low temperatures shows a $\sim -T^{1/2}$ -dependence. Up till now, the $\rho(T) \sim -T^{1/2}$ dependence has been found only in weakly disordered three-dimensional metallic conductors (Metglas alloys) [14] and not in single-crystalline specimens.
- (ii) Rozenberg *et al* mention in their comment that for $T < T_{min}$ the resistivity is flattened and the minimum degrades upon increasing the external magnetic field and will disappear at $H = H_{cr}$. This assertion is in contradiction with the above-mentioned results of Kumar and collaborators (especially points (b) and (d)) [7].
- (iii) After a careful study of figure 2 in [4] one can clearly see that the resistivity of polycrystalline $\text{La}_{0.80}\text{Sr}_{0.20}\text{MnO}_3$ (ceramic in Auslender's notation) at very high fields of $H = 10$ and 13.6 T respectively displays a minimum and a notable upturn at low temperatures. It seems that even these high magnetic fields do not shift, flatten or destroy the minimum. Unfortunately, their experimental data on the upturn resistivity are very poor, so no conclusion can be drawn regarding its temperature dependence at $T < T_{min}$.

In conclusion, these contradictory interpretations make this effect very interesting and in our opinion need further precise experimental investigation and theoretical work.

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