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COMMENT

Comment on 'Low-temperature transport properties of non-stoichiometric $La_{0.95-x}Sr_xMnO_3$'

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Abstract

In a recent paper Michalopoulou *et al* (Michalopolou A, Syskakis E and Papastaikoudis C 2001 *J. Phys.: Condens. Matter* **13** 11 615) reported the measurements of electrical resistivity and specific heat at zero magnetic field carried out on polycrystalline non-stoichiometric $La_{0.95-x}Sr_xMnO_3$ manganites. In particular, the authors attributed the low temperature (LT) behaviour of resistivity (shallow minimum and slight upturn at lowest temperatures) to three-dimensional electron–electron interaction enhanced by disorder, using results of numerical fittings of resistivity versus temperature dependences in the interval 4.2–40 K. It is shown in this comment that such analysis may be not valid for polycrystalline manganites where relatively strong grain boundary effects might mask a weak contribution of quantum effects to the LT resistivity. The crucial test of applicability of the theory of quantum corrections to conductivity in this case is the resistive measurements under non-zero magnetic field.

Michalopoulou *et al* presented in their recent paper [1] data on electrical resistivity and specific heat measured at zero external magnetic field (H) on polycrystalline non-stoichiometric $La_{0.95-x}Sr_xMnO_3$ manganites in the doping region $0 \leqslant x \leqslant 0.3$. Using numerical fittings of resistivity (ρ) versus temperature (T) dependences in the interval 4.2–40 K, authors claim that the low temperature (LT) behaviour of resistivity (shallow minimum and slight upturn at lowest T) may be accounted for by three-dimensional electron–electron interaction enhanced by disorder [2], as suggested previously in [3–5] for polycrystalline manganites. At the same time, applicability of the theory of quantum corrections to conductivity (QCC) [2] was checked in detail for single crystals and ceramics of different doped manganites in our recent papers [6, 7]. Let us note briefly the main results reported in [6, 7] and compare them with those of the paper commented upon [1].

(i) It should be emphasized that QCC theory [2] is a 'bulk' one, describing influence of quantum effects such as electron–electron interaction and weak localization on LT conductivity of bulk (single crystalline) metals and compounds with metallic properties. Thus, one should be extremely careful using the QCC model for description of LT resistivity of polycrystalline samples.

(ii) In particular, formal numerical fitting of the zero-field resistivity upturn at lowest T with a $\sim -T^{1/2}$-dependence alone is not sufficient for single-valued verification of the applicability of the QCC theory [6]. A crucial test of any theoretical model in this case is the influence of non-zero H on the LT minimum of ρ.

(iii) It was shown in [6] and tested additionally in [7] that the above minimum (observed at $T_{min} \sim$ 20–30 K) is flattened and vanishes under moderate external H of about 1.5 and 10 T in $La_{0.5}Pb_{0.5}MnO_3$ (LPMO) and $La_{0.8}Sr_{0.2}MnO_3$ (LSMO) ceramics, respectively. Such behaviour strongly contradicts the prediction of QCC, according to which the LT minimum of ρ persists and is affected very weakly by H of about 10 T [6].

(iv) An alternative model of carrier tunnelling between antiferromagnetically coupled grains, taking into account grain boundary (GB) effects in polycrystalline manganites, provides a fairly good qualitative description of the above (point (iii)) phenomenon [6]. But notable differences are observed in LT conductivity for relatively 'poor' LPMO and 'good' LSMO ceramics (mean grain size is smaller by an order of magnitude and residual ρ is higher by two orders of magnitude in the former sample compared to the latter one). Firstly, the critical H that makes the 'GB' minimum vanish in LSMO is higher than that for LPMO (point (iii)) and, secondly, an additional field independent very weak LT minimum of ρ that was previously masked by the relatively strong 'GB' minimum appears at $H \geqslant 10$ T in LSMO, while it is absent for LPMO for H higher than the critical value [7].

(v) This LT minimum of ρ in the LSMO polycrystalline sample, as well as the slight H-independent upturn of resistivity observed in a single crystal of LSMO at liquid helium temperatures were attributed to bulklike LT conductivity governed by QCC [7].

We contend that the above points (i)–(v) are extremely important for the following comments on results of Michalopoulou *et al* [1].

(1) In general, Michalopoulou *et al* did not present in the paper commented upon [1] any systematical data on conductive and magnetic properties i.e. Curie points, temperatures of metal–insulator transition etc of the investigated $La_{0.95-x}Sr_xMnO_3$ system. However, it was pointed out that the parent compound $La_{0.95}MnO_3$, as well as $La_{0.85}Sr_{0.1}MnO_3$, is an insulator (the latter sample may be attributed to an inhomogeneous insulator with phase-separated ground state—see pp 11 617 and 11 620 in [1]). Temperature-dependent percolation of metal-like domains within an insulating matrix may be a possible nature of the LT minimum of ρ observed in $La_{0.85}Sr_{0.1}MnO_3$—figure 2(a) in [1]. This means that QCC theory in principle could not be used for analysis of LT conductivity in $La_{0.85}Sr_{0.1}MnO_3$—see point (i)—and physical mechanisms completely different from QCC may govern the LT conductivity of this sample (see, for example [8, 9]).

(2) Nevertheless, LT upturns of ρ observed for $La_{0.85}Sr_{0.1}MnO_3$, as well as for $La_{0.75}Sr_{0.2}MnO_3$ and $La_{0.65}Sr_{0.3}MnO_3$ are pretty well fitted by a $\sim -T^{1/2}$-dependence—figure 2 in [1]. This fact well illustrates and strongly supports point (ii) on the insufficiency of only numerical fitting of the ρ versus T dependence at $H = 0$ in arguing for the applicability of the QCC model.

(3) It is impossible, of course, to disclaim in principle the existence of a QCC-like contribution to the LT conductivity of metallic-like polycrystalline $La_{0.75}Sr_{0.2}MnO_3$ and $La_{0.65}Sr_{0.3}MnO_3$ samples. But, taking into account our previous results [6, 7] and very

recent data by Roy *et al* [10], it is possible to assume that the experimentally observed LT minimum of ρ at $H = 0$—figures 2(b), (c) in [1]—originates from GB effects (points (iii), (iv)) in the above ceramics. Such supposition is confirmed by about the same values of $T_{min} \sim 20$ K and the LT upturn of $\rho \sim 1\%$ (compare figures 2(b), (c) in [1] and data [6, 7, 10]) observed at $H = 0$ on different polycrystals of doped manganites. It is interesting to note that in insulator-like $La_{0.85}Sr_{0.1}MnO_3$ ceramic such an upturn is notably stronger (about 4%)—figure 2(a) in [1].

(4) The crucial test of the validity of interpretation proposed by Michalopoulou *et al* is measurement of LT ρ versus T dependences under external H of a few Tesla—point (ii). It may be predicted certainly that the GB-like minimum of resistivity will be flattened and vanish in such a field [6, 7, 10] and the almost H-independent very weak bulklike minimum will appear (if it exists) [7]. Taking into account the relatively low values of residual resistivity of $La_{0.75}Sr_{0.2}MnO_3$ and $La_{0.65}Sr_{0.3}MnO_3$ samples, the temperature of such a bulklike minimum (described by the QCC model) may be estimated as $T_{min} \leqslant 10$ K [7].

(5) Finally, let us note that Matthiessen's rule used in [1] for determination of the possible mechanism of inelastic contribution to the conductivity at $T > T_{min}$ is valid only for metallic-like systems. Thus, the value obtained for the insulating-like $La_{0.85}Sr_{0.1}MnO_3$ must be excluded from figure 3 in [1]. The simplest way to choose between usual electron–electron and unconventional Furukawa scatterings is measuring and fitting of resistivity through an extended interval of temperature above T_{min} (in the paper commented upon [1] such fittings were done for a range ΔT of about 10–20 K only).

To conclude, the simplified analysis of experimental data presented by Michalopoulou *et al* in [1] based on fitting of LT resistivity versus temperature dependences measured only at zero magnetic field is absolutely insufficient for verification of the applicability of the QCC model (as well as in previous analogous attempts [3–5]). At the same time, analysis of specific heat data (typical bulk property) presented in [1] seems plausible enough.

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