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Low temperature resistivity anomalies in Pr-based nano-manganites

Proloy T. Das, A.Taraphder and T. K. Nath
Department of Physics, Indian Institute of Technology Kharagpur,
West Bengal - 721302, India
dasproloy2007@gmail.com ; tnath@phy.iitkgp.ernet.in

ABSTRACT

The low-temperature electronic transport behavior of under doped polycrystalline $Pr_{0.8}Sr_{0.2}MnO_3$ (PSMO) manganite nanoparticles (down to 40 nm), have been investigated in the presence of applied external magnetic fields (H_{ext}) and a distinct resistivity minimum (ρ_{min}) is observed below 50 K for each PSMO sample. It has been found that both depth of ρ_{min}, and temperature of resistivity minima ($T_{\rho_{\min}}$) values enhance with increase of H_{ext}. Considering various possibilities like Coulomb blockade theory, electron-electron interaction, phase separation, Kondo mechanism, we conclude that occurrence of low temperature resistivity anomalies ($<T_{\rho_{\min}}$) in PSMO manganite system is presumably due to a combined effect of electron-electron interaction ($\sim T^{1/2}$) and 3D weak localization (WL) mechanism. The proposed model can explain spin dependent scattering phenomena in disorder background of correlated manganite system and behavior of various fit parameters responsible for low temperature resistivity anomalies under external magnetic fields.

Keywords

Manganites, Resistivity minima, electron-electron interaction, weak localization

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I INTRODUCTION

Colossal magneto-resistive oxides (CMR) [1] have been studied quite intensely since last a few decades due to their unique electronic and magnetic properties which have a technologically potential importance in sensor or magnetic memory device applications. Out of many fascinating properties, low temperature resistivity anomaly is an interesting feature of CMR manganites and in recent years it has been well reviewed by several investigators [2-10] with various plausible origins: such as Quantum interference effect [3, 7], spin dependent scattering through multi resistive phase [4], Kondo-like mechanism [5, 6], and Coulomb blockade theory [9, 10], and so on. However, proper analysis and interpretation of the observed low temperature resistivity minima (ρ_{min}) in the manganite system is still under discussion. The interesting question over decades is to correlate this phenomenon with Coulomb blockade type theory or modified Kondo like transport mechanism or competition between electron-electron inelastic scattering like processes and 3D weak localization [WL] theory [11, 12] including spin polarization, grain boundary tunneling in the polycrystalline manganites [13].

Motivation of this present work is to obtain new insight into the low temperature resistivity anomaly in under doped PSMO nano-manganite system considering a combined effect of electron-electron interaction and 3D weak localization (WL). The role of spin dependent scattering on low temperature resistivity anomaly has also been investigated.

II Experimental details

The single phase PSMO samples were synthesized by chemical pyrophoric reaction route. The required quantities of respective oxide or carbonate powders were mixed and sintered at different sintering temperatures of 850 ^{0}C, 950 ^{0}C, and 1050 ^{0}C in air to obtain 40 nm (PSMO 850), 50 nm (PSMO 950), and 58 nm (PSMO 1050) nanometric grain size samples, respectively. The microstructural and surface topography of PSMO samples were already reported elsewhere [14]. Temperature dependent resistivity measurement (5 - 300 K) has been carried out in presence of 0, 5, and 9 T magnetic fields using standard four probe dc techniques in a CFM–VTI 9 T superconducting magnet system (Cryogenic Limited, U.K.). During the measurement, temperature stability was better than ± 50 mK.

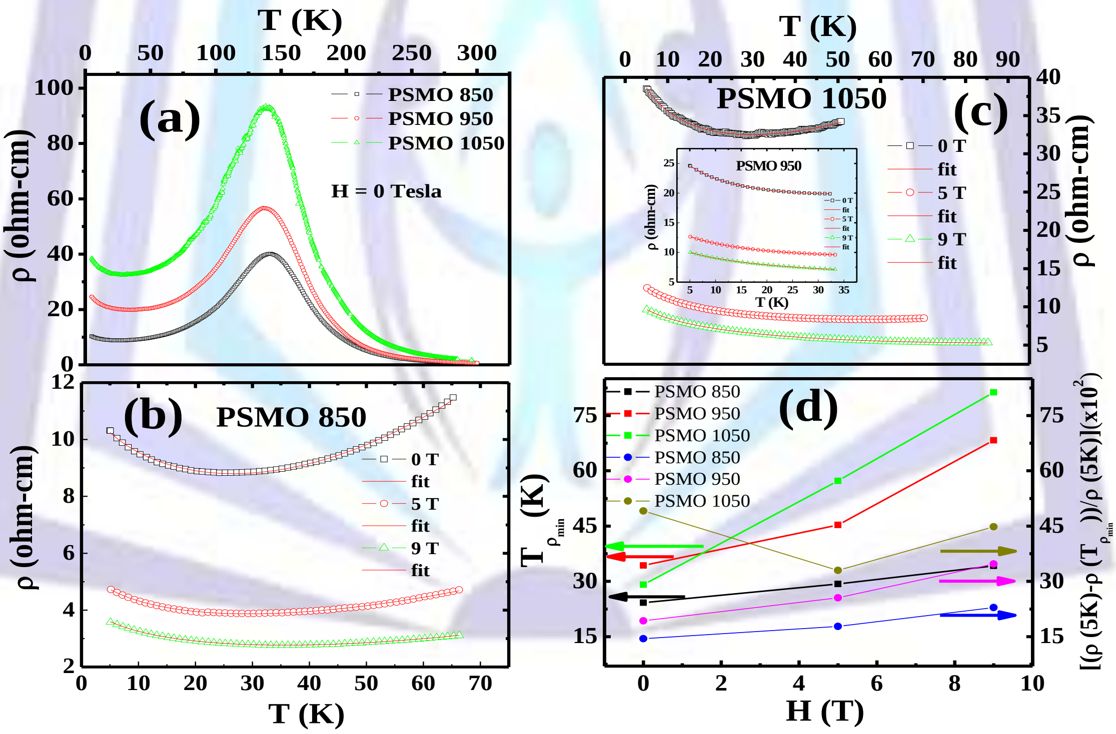

Figure 1 (color online): (a) Temperature dependence of resistivity of PSMO samples at 0 T; (b), and (c) Fits of low temperature resistivity data of PSMO samples recorded at various magnetic fields; (d) Variation of $T_{\rho_{min}}$, and depth of resistivity minima with various applied magnetic fields.

III Results and Discussion

High resolution zero field *ρ(T)* data for each PSMO sample in the range of 5 - 300 K is shown in Fig. 1(a). It has been found that metal-insulator transition (MIT) appears for all PSMO samples around 140 K and shows ferromagnetic nature < 150 K [14]. Each sample exhibit distinct ρ_{min} along with resistivity upturn below MIT (< 50 K) at various external magnetic field (H_{ext}) shown in Fig. 1(b) and 1(c), respectively. Figure 1(d) depicts that resistivity minima of all samples are not disappeared after applying of H_{ext} even upto 9 T and $T_{\rho_{min}}$ shifts toward high temperature regime for all samples. To explore the possible mechanism of it, we have fitted observed low temperature resistivity data with various plausible models such as Coulomb blockade, phase separation theory, Kondo-like mechanism, electron-electron interaction mechanism etc. It has been found that, Coulomb blockade theory of electrostatic origin fits well with our lowest nanometric samples (PSMO 850, PSMO 950) compared to PSMO 1050. It is also found from Fig. 1 (a) that, PSMO 1050 exhibit higher resistivity value compared to other two samples which signifies applicability of Coulomb blockade theory is restricted within lower nanometric grain size or in very low temperature regime < 5 K [9,15]. In connection, any drastic change in ρ(T) has not been observed even after changing the sample current during measurement which rules out the possibility of tunneling between multi resistive phase [4].

Kondo mechanism [16], originating from the interaction of localized spins of magnetic impurities and conducting electrons in dilute magnetic alloys, generally occurs at low temperature (typically below 6 K) whereas for PSMO systems $T_{\rho_{\min}}$ is observed below 50 K. Moreover, it is noticeable that depth of ρ_{min} does not disappear at different external magnetic fields even upto 9 T, in contrast to the feature generally observed in Kondo scattering mechanism. Recently, some researchers [5, 6] have attempted to explain the observed this resistive anomaly with concept of Kondo-like mechanism. To interpret the physical mechanism, we have fitted the low temperature ρ(T) data considering Kondo like mechanism and elastic electron-electron interaction together in the same frame work and it is given by the relation,

$$\rho(T) = \rho_0 - \rho_S \ln(T) - \rho_{e^/} T^{1/2} + \rho_e T^2 + \rho_p T^5 \text{..............(1)}$$

where ρ_0 is the residual resistivity, arising from elastic scattering like electron-impurity scattering and crystal imperfection, $\rho_s \ln(T)$ is ascribed to Kondo like scattering. The term $\rho_e^{/}T^{1/2}$ arises for elastic electron-electron interaction. $\rho_e T^2$ term is due to inelastic scattering (electron-electron/electron-magnon) contribution, and the $\rho_p T^5$ term is from electron-phonon interaction originated from Bloch-Gruneisen integral term [17]. After the fiiting procudure with Eq. (1) for all samples, it has been observed that experimental data is not well fitted in entire low temperature regime with eq. (1) and rules out the possibility of Kondo mechanism in the studied PSMO system. Usually, occurence of low temperature resistivity anomaly in these strongly correalted systems appears due to electron-electron interaction and disorder presents in the said system [2,13]. In the present case, we found that depth of resistivity minima of all PSMO samples, shift toward high value side with induction of H_{ext} which is a distinct signature of electron-electron interaction and WL mechanisnm [18, 19]. For better understanding of the low temperature resistivity upturn in the present PSMO system, a combined model of electron-electron interaction, Magneto-resistance (MR) and 3D weak localization contributions [2, 20] can be proposed in the following expression,

$$\rho(T,H) = \rho_0 + \rho_m(T,H) - \rho_0{}^2[\sigma_{ee}(T,H) + \sigma_{WL}(T,H)] + \rho_n(T,H)T^n \text{...............(2)}$$

Where ρ_0 is residual resistivity, $\rho_m(T,H)$ arises due to magneto resistance(MR) contribution from electron-magnon scattering, $\sigma_{ee}(T,H), \sigma_{WL}(T,H)$ are the conductivities for electron–electron interaction and WL contributions, respectively. Here $\rho_n(T,H)T^n$ is the contribution due to inelastic scattering effect. The important issue here is to find out the applicability of WL mechanism (interference of two time reversal partial electron wave functions) in the studied material. In the present case, all PSMO samples show high resistivity values ~30-40 Ω-cm at 5 K which is quite greater than Motts's maximum metallic resistivity limit ~ 10 mΩ-cm which also favor the electron-electron elastic scattering mechanism (appears due to columbic interaction of charge carriers and weak localization) [21-23]. Recently, Guo et al. [24] predicted that- (i) electron interference effect can be destroyed under strong magnetic field in ferromagnetic ground state [25] and, (ii) WL theory is valid in reduced dimension (<20 nm) [26], however in contradiction, Burgy et al. suggested that presence of any kind of inhomogeneity (structural/magnetic/electronic) is quite conventional in any doped oxide system, eventually it is the inherent property of that doped system even in the best crystals[27]. In addition, it is also proposed that temperature dependency of ferromagnetic clusters in strongly correlated electron system can be interpreted with disorder theory which indicates the probability of existence of WL mechanism in that system as well[28]. Usually in electronic transport of manganite systems WL part remains overshadowed by dominating electron-electron interaction part [3] as WL contribution is much less than electron-electron interaction counterpart. To solve the issue in the studied PSMO system, we have checked contributions of electron-electron interaction and WL part in successive manner.

Firstly, we have considered elastic and inelastic scattering contribution only by dropping down and neglecting minor contribution of WL and MR parts from eqn. (2) which can be simplified below MIT as,

$$\rho(T,H) = \rho_0 - \rho_{e^/} T^{\frac{1}{2}} + \rho_n(T,H)T^n \text{..................(3)}$$

Here all the parameters have their ususal physical significance [5,6]. Coefficient $\rho_e^{/}$ is for electron-electron interference effect ~ $D^{-1/2}$ (diffusion constant), and n is inelastic scattering exponent for possible inelastic interaction. We have also tried to fit our experimental ρ(T,H) data including electron-phonon scattering term considering T^2 dependence (to distinguish separately the inelastic contribution of electron-electron, electron-magnon, and electron-phonon interactions) using Eq. (3). It can be seen that the data fit well with Eq.(3), although the coefficients are randomly varied. In the low temperature magneto-transport measurement, any positive MR in the studied PSMO system has not been observed [14] however, it was expected from electron-electron interaction. It is found that, measured experimental data shows an excellent agreement with electron-electron interaction model as shown in Fig. 1. The extracted fitting parameters (ρ_0, ρ_e. ρ_n, n, and χ^2) of all samples employing Eq. (3) at different magnetic fields are summarized in Table I.

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Table I: Various coefficients of fitted parameters using Eq. (3), and the quality of fit, χ^2 values for PSMO samples in presence of different H_{ext}.

Sample name PSMO	H_{ex} (T)	ρ_0 (Ω-cm)	$\rho_e^{/}$ (Ω-cm /$K^{1/2}$) (10^{-2})	ρ_n (Ω-cm/K^n) (10^{-2})	n	χ^2 (10^{-5})
	0	12.27	93.4	0.40	1.780	1.11
850	5	6.05	64.3	1.42	1.333	22.35
	9	4.67	52.2	1.22	1.281	23.36
	0	30.04	262.0	2.53	1.509	8.90
950	5	15.98	159.1	2.97	1.294	44.95
	9	13.20	167.9	12.8	0.955	96.27
	0	50.80	717.0	86.0	0.939	7.60
1050	5	17.91	368.8	83.6	0.763	22.94
	9	13.63	263.8	58.6	0.744	16.97

From the Table-I, it is quiet obvious that all of the fitting parameters of Eq. (3) carries a strong dependence with external magnetic field, H_{ext}. Coefficients like ρ_0, ρ_e' are suppressed gradually with H_{ext} as expected. Experimental fit value of ρ_n is found to be less at higher magnetic field (9T) for PSMO 850 and PSMO 950 samples wheras the value of ρ_n was reversed in case of PSMO1050. It is known that for metallic conductors, residual resistivity, ρ_0 does not depend on temperature as well as magnetic fields, however in present system a nonlinear nature with sample sintering temperature has been observed which shows a decreasing nature with increase of H_{ext}. The exact reason of it is not clear, might be due to weak disorder of the present system. Similar kind of nature has also been observed for ρ_e and ρ_n with sintering temperature variation which can be attributed to magnetic scattering and inelastic electron-electron interaction of charge carriers. It is also found that for all PSMO samples, $\rho_e^{/}$ decreases gradually with H_{ext} and depth of ρ_{min} increases with H_{ext}, however for PSMO 1050, $\rho_e^{/}$ has higher value than other two. The gradually decrease of n with H_{ext} reflects the suppression of inelastic scattering, and increasing of $T_{\rho_{\min}}$ with applied fields [29]. In the proposed electron-electron intercation model, the goodness of fit, χ^2 has been found quiet less ~10^{-5} whereas the same χ^2 was ~10^{-3} for Coulomb blockade and Kondo like models respectively which also signifies the applicability of electron-electron interaction model in the present PSMO system. PSMO 850 and PSMO 950 nanomanganites exhibit a large grain surface disorder and spin dependent tunneling through the inter-grain resistive path as suface to voleme ratio increases whereas in PSMO 1050 sample, charge carriers are more localized whic makes it more resistive[14]. Recently, the origin of low temperature resistivity anomaly in $La_{2/3}Sr_{1/3}MnO_3$-ZrO_2 system has been explored with a combined frame work of weak localization and electron-electron interaction [30]. In the present PSMO system, similar kind of correlation of the aforesaid model has been found.

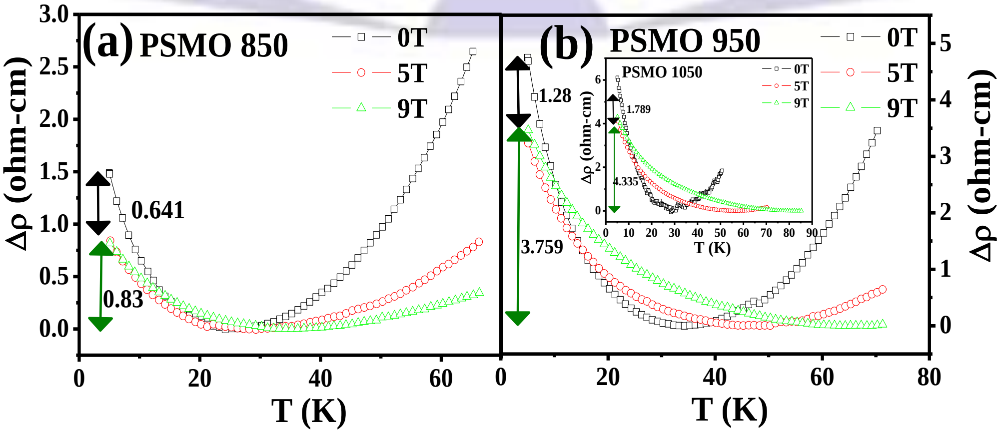

Figure 2 (color online): (a-b) $\Delta\rho(T)$ (= $\rho(T)$- ρ_{min}) variation with temperature for all PSMO samples; black arrow: field dependent regime and deep green arrow: field independent regime.

Secondly, to find out MR contribution in low temperature ρ(T) data below ρ_{min} of the studied system an approach has been taken by considering Δρ (T) = ρ(T) - ρ_{min} at different applied magnetic fields (0T, 5T, and 9T) as shown in Fig. 2. It was found that $\rho_{H_{min}}$ slowly varies with temperature for all samples which turn out that effect of MR contribution is minimal compared to raw resistivity data. Field dependent MR part increases gradually with grain size variation (as marked by black arrow: ~0.641 for PSMO 850, ~1.280 for PSMO 950 and ~1.789 for PSMO 1050 samples). Thus we see here WL part, $\rho_{WL} = \Delta\rho_{0Tesla} - \Delta\rho_{9Tesla}$ decreases with size reduction of PSMO nanomanganite system.Variation of field independent part of MR, $\Delta\rho_{9Tesla}$(T) in grain size modulation, (as marked by deep green arrow: ~0.830, ~3.759, and ~4.335 for PSMO 850, 950, and 1050 samples respectively) depicts a consistent behavior with the earlier report on $La_{2/3}Sr_{1/3}MnO_3$ system [20]. Figure 2(a) and 2(b) depicts that $\Delta\rho_{5Tesla}$(T) and $\Delta\rho_{9Tesla}$ (T) data are almost superposed on each other confirming the existence of WL contribution which was totally suppressed by H_{ext} at lower temperature [31]. In the dc magnetization measurement at 500 Oe, a moderate bifurcation in low temperature regime(<<T_C) between field cooling (FC) and zero field cooling (ZFC) data [14] has been observed which could be a direct evidence of disorder presents in these kind of strongly correlated systems and measures the degree of inhomogenity, disorderness, and presence of glassy states of said mangaite systems[15].Usually spin dependent disorderness and scattering in low temperature regime of these corelated systems come from weak disorder and is the most dominant factor for occurence of low temperture resistivity upturn.

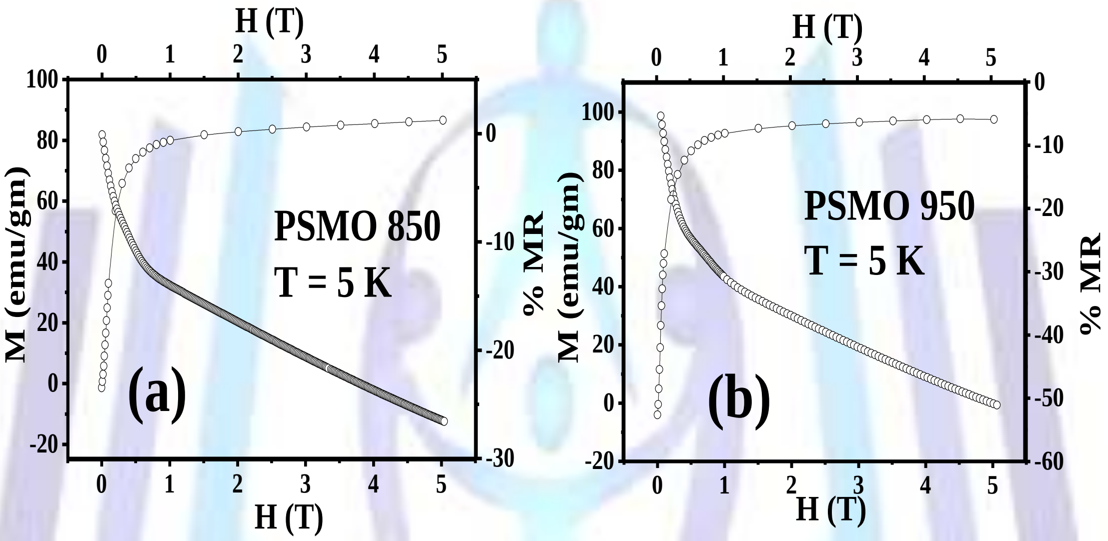

Figure 3 (color online): (a-b) Simultaneous resistivity and magnetization plot with H_{ext} for PSMO 850 and PSMO 950 samples

In Fig. 3, a simultaneous dependency of MR and magnetization with tuning of H_{ext} at 5 K are shown from which it is evident that, in lower field both MR and DC-magnetization behaviour show a nonlinear dependency (change of slopes in different domain), however in higher field side (> 1T) same exhibits an almost linear dependence behaviour. In addition, no satuartaion nature has been observed for PSMO 850 and PSMO 950 samples in higher magnetic field side (>1T) which is a well known feature of any manganite system, arises from system disorderness. However, with enhancement of H_{ext} disorderness presents in the studied system was suppressed as shown in Fig. 2 which kind of feature was also found in polycrystalline $La_{2/3}Sr_{1/3}MnO_3$ system[6].

IV Conclusions

In summary, we have investigated the possible physical origin of resistivity upturn behaviour below $T_{\rho_{min}}$ under various magnetic fields for the $Pr_{0.8}Sr_{0.2}MnO_3$ manganite system with different conventional models. Results depict that $T_{\rho_{min}}$ of all samples were shifted toward to higher value side with enhancement of H_{ext} and depth of ρ_{min} was not smeared out under presence of H_{ext}. Considering most probable possibilitis with various fitting procedure, we conclude that in the present PSMO system both electron-electron interaction and 3D WL mechanism both have significant contributions for occurenece of resistivity minima <50 K however, electron-electron interaction part holds the dominating role. However, we believe that further theoretical and experimental invesigations are also needed to explore low temperature resistivity anomalies of these phase separeted metallic manganite systems under a common framework of electron-electron interaction and 3D WL model.

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