Pulsed versus DC I-V characteristics of resistive manganites

B. Fisher, J. Genossar, K. B. Chashka, L. Patlagan, and G. M. Reisner

Physics Department and Crown Center for Superconductivity, Technion, Haifa 32000, Israel.

(Dated: August 25, 2018)

Abstract

We report on pulsed and DC I-V characteristics of polycrystalline samples of three charge-ordered manganites, Pr$_{2/3}$Ca$_{1/3}$MnO$_3$, Pr$_{1/2}$Ca$_{1/2}$MnO$_3$, Bi$_{1/2}$Sr$_{1/2}$MnO$_3$ and of a double-perovskite Sr$_2$MnReO$_6$, in a temperature range where their ohmic resistivity obeys the Efros-Shklovskii variable range hopping relation. For all samples, the DC I(V) exhibits at high currents negative differential resistance and hysteresis, which mask a perfectly ohmic or a moderately nonohmic conductivity obtained by pulsed measurements. This demonstrates that the widely used DC I-V measurements are usually misleading.
Charge ordering (CO), often associated with spin (antiferromagnetic (AF)) ordering and its collapse in manganite perovskites, have been the subject of intense investigations over the past decade. The electric field induced current switching in a CO Pr$_{0.7}$Ca$_{0.3}$MnO$_3$ single crystal, at temperatures below the CO transition temperature ($T_{CO}$), attracted great interest. Investigations of the non-linear conductivity of single crystals, films and polycrystalline samples of different CO manganites were reported by many groups and gave rise to several ad-hoc interpretations. (For a short review see Ref. 4). All these I-V characteristics were obtained with DC currents. In most studies, Joule heating was ruled out by calculating the expected temperature increment ($\Delta T$) and/or by monitoring it close to the samples’ surface. However, both the calculations and the measurements showing $\Delta T$ to be negligible were misleading, being based on the assumption that the current flow throughout the crosssection of the sample and thus its temperature are homogeneous. An elaborate investigation on polycrystalline samples of Pr$_{0.8}$Ca$_{0.2}$MnO$_3$ (with no CO but exhibiting nonlinear conductivity) has shown that the internal thermal gradient caused by Joule heating is the origin of the I(V) nonlinearity. Thermal filaments in switching samples of other materials have been visually observed in the past.

An old but lately neglected technique for preventing and confirming absence of Joule heating errors in high E-field measurements, consists of applying pulsed currents and following the time dependence of the response on an oscilloscope. For short rise-time square pulses, Joule heating is negligible as long as the response is independent of time.

Here we report on a comparative study of DC and pulsed I-V characteristics of polycrystalline samples of Pr$_{1-x}$Ca$_x$MnO$_3$ with $x = 1/3$ (PCMO(1/3)) and $x = 1/2$ (PCMO(1/2)), Bi$_{1/2}$Sr$_{1/2}$MnO$_3$ (BSMO(1/2)) and Sr$_2$MnReO$_6$ (SMRO). PCMO(1/3), PCMO(1/2), and BSMO(1/2) are CO-AF manganites ($T_{CO}$=240, 220 and 450 K and $T_N$=180, 150 and 150 K, respectively) while SMRO is a double-perovskite that undergoes a transition to insulating ferrimagnet at $T_N$=120 K. The resistivity of all these systems follows, over wide ranges of $T$, the Efros-Shklovskii variable range hopping (ES-VRH) relation $\rho(T) \propto \exp(T_o/T)^{1/2}$. We show that the pulsed $J$ - $E$ characteristics ($J$ - current density and $E$ - electric field) are linear for most samples or weakly nonlinear for a few, up to fields around $10^3$ V/cm, while those obtained with DC measurements are nonlinear and exhibit negative differential resistance (NDR) at much lower fields.

The samples were prepared according to protocols taken from Ref. 10 for PCMO, Ref. 9 for
BSMO and Ref. 7 for SMRO. X-ray diffraction (XRD) measurements have been performed using a Siemens D5000 powder diffractometer with CuKα radiation. None of the XRD powder patterns showed foreign phases.

Most I-V characteristics were measured by the four-probe technique (1 and 4 - the current contacts, 2 and 3 - the voltage probes). The voltage probe separations ranged between 0.9 to 1.6 mm, about 1/3 of the total lengths of the samples. To extend the pulsed measurements to higher fields and lower temperatures we used also short samples of about 1 mm length. They were used only when the two-probe ρ(T), agreed with the four-probe one (within the uncertainty of the dimensions). The agreement was very good for the BSMO(1/2) and SMRO samples at all temperatures and for PCMO samples below ~ 100 K.

DC I-V characteristics were measured over many orders of magnitude up to the current runaway in the NDR regime. Under this restriction, the measured surface ΔT never exceeded a fraction of a degree.

For pulsed measurements, single pulses in the millisecond range were applied from a Keithley 237 high voltage source; its maximal voltage was 150V for currents up to 10mA. Using the two channels of a Tektronix 2221A digital storage oscilloscope we recorded simultaneously, as function of time the voltage drops between the ground and pairs of voltage probes. For each pair (V₀₂, V₀₃ and V₀₁, V₀₄) the measurement was repeated four times in order to average out the small fluctuations. V₀₁, on a small resistance in series with the sample and V₁₄ = V₀₄ − V₀₁ were used to check the current and the quality of the current contacts, respectively. A V₁₄/V₂₃ ratio, independent of E and T and close to that expected from the dimensions, is an additional test for the use of two-probe samples.

Fig. 1 shows ρ(T) of PCMO(1/2), PCMO(1/3), BSMO(1/2) and SMRO plotted on a semilog graph as function of T⁻¹/₂. The solid lines represent the relation \( \ln \rho = \rho_o \exp(T_o/T)^{1/2} \) that fits very well the data over at least four orders of magnitude. Unexpectedly, the plots for BSMO(1/2) and SMRO and sections of the PCMO(1/3) and PCMO(1/2) plots are almost parallel straight lines. Their slopes correspond to values of T₀ within the range between \( 5.1 \times 10^4 \) for PCMO(1/2) to \( 5.7 \times 10^4 \), for BSMO(1/2). According to the ES-VRH model, \( e_r a = 2.8e^2/(4\pi\epsilon_o k_B T_o) \), where \( a \) is the localization length and \( e_r \) - the relative dielectric constant. The above values of T₀ correspond to \( e_r a \approx 0.9 \text{nm} \); an atomic length-scale of \( a \), seems reasonable.

Fig. 2(a) shows the pulsed (symbols) and the DC (solid lines) J-E characteristics mea-
sured at various temperatures on PCMO(1/3), down to 80 K on a four-probe sample, and at 60 and 70 K on a two-probe one. The pulsed four-probe $J-E$ characteristics are ohmic up to $E \sim 800 \text{ V/cm}$ while those obtained for the two-probe sample are non-ohmic. For both two-probe plots $J/J_{ohm}$ ($J_{ohm}$ - the extrapolated ohmic current) is 1.85 at the maximal field ($E = 1480 \text{ V/cm}$). In Fig. 2(b) the pulsed two-probe $J-E$ characteristics of a PCMO(1/2) sample are perfectly ohmic.

The DC $J(E)$ plots in Fig. 2, for both PCMO(1/3) and PCMO(1/2), are linear over several orders of magnitude of $E$ but become strongly nonlinear, exhibit NDR and hysteresis at moderate fields (see insets for better resolution). The field at the turnover into the NDR regime increases with decreasing $T$. The main features of these DC characteristics are as calculated by models of self-heating.$^{12}$

Fig. 3 shows the pulsed and the DC $J-E$ characteristics of BSMO(1/2) samples, measured at various temperatures, down to 90 K on a four-probe sample and for 80 K on a two-probe sample. All pulsed $J-E$ characteristics are ohmic, while the DC $J(E)$ are similar to those of the PCMO samples.

Fig. 4 shows the pulsed and the DC $J-E$ characteristics measured on SMRO samples, down to 120 K on a four-probe sample, and at 100 K, on a two-probe sample. The pulsed $J-E$ characteristics are nonlinear at fields as low as $\sim 100 \text{ V/cm}$. The non-linearity increases with decreasing $T$ and increasing $E$. DC and pulsed $J(E)$ are identical in the low-field nonlinear regime but diverge at higher fields. For the pulsed measurements, $J/J_{ohm}$ is 2.7 at $T = 120 \text{ K}$ and $E = 580 \text{ V/cm}$ and, 4.6 at $T = 100 \text{ K}$ and $E = 1560 \text{ V/cm}$. After separation, the DC $J(E)$ increase much faster towards the NDR regime. The nonlinearity in the pulsed $J(E)$ data for both types of SMRO samples, indicates that in the case of PCMO(3) two-probe sample it may not be an artifact of this configuration. Nonlinear $J(E)$ determined by the parameter $\epsilon E a / k_B T$ is expected in case of hopping conductivity.$^{11}$ For $T \geq 100 \text{ K}$ and $a \approx 1 \text{ nm}$ (see above), one would expect nonlinear $J(E)$ at much higher fields than reported here. On the other hand, the various models suggested for the nonlinear conductivity in CO manganites are irrelevant for SMRO.

The absence of broad band and telegraph noise that would be associated with CO de-pinning, filament formation or other instabilities is confirmed by the flatness (within three digit resolution, over several milliseconds) of the observed voltage drops.

In summary, for all samples investigated here, CO manganites and a resistive double-
perovskite, the DC I-V characteristics mask a perfect linearity or a moderate nonlinearity of $J(E)$ observed by pulsed measurements. This demonstrates that the widely used DC I-V measurements are usually misleading.

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FIG. 1: Semilog plot of $\rho$ versus $T^{-1/2}$ for $\text{Pr}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$, $\text{Pr}_{1/2}\text{Ca}_{1/2}\text{MnO}_3$, $\text{Bi}_{1/2}\text{Sr}_{1/2}\text{MnO}_3$, $\text{Sr}_2\text{MnReO}_6$. Solid lines represent the relation $\rho = \rho_o \exp(T_o/T)^{1/2}$ that fits each set of experimental data over at least four orders of magnitude of $\rho$.

FIG. 2: $J$-$E$ characteristics of (a) $\text{Pr}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$ samples measured in four-probe, and two-probe configurations, down to 90 K and below, respectively and (b) on a $\text{Pr}_{1/2}\text{Ca}_{1/2}\text{MnO}_3$ sample, in a two-probe configuration. Symbols for pulses and solid lines for DC. Dashed lines in (a) represent the extrapolated ohmic current - $J_o$. Insets show $J(E)$ on linear scale for 80 K in (a) and for 60 K in (b).

FIG. 3: $J$-$E$ characteristics of $\text{Bi}_{1/2}\text{Sr}_{1/2}\text{MnO}_3$ samples (in four-probe, or two-probe configurations, down to 90 K or at 80 K, respectively). Symbols for pulses and solid lines for DC. The inset shows the plots for 80 K on a linear scale.

FIG. 4: $J$-$E$ characteristics of $\text{Sr}_2\text{MnReO}_6$ samples (in four-probe, or two-probe configuration, down to 120 K or at 100 K, respectively). Dashed lines represent the ohmic currents. Symbols for pulses and solid lines for DC. The inset shows the plots for 100 K on a linear scale.
Fig. 1

$\rho$ (\(\Omega\) cm)

$T^{-1/2}$ (K\(^{-1/2}\))

- SMRO: \(-5.66 \times 10^4\)
- BSMO(1/2): \(-5.71 \times 10^4\)
- PCMO(1/3): \(-5.57 \times 10^4\)
- PCMO(1/2): \(-5.11 \times 10^4\)
Fig. 2
Fig. 3
Fig. 4