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Room temperature giant magnetoimpedance in $La_{0.7}Ba_{0.15}Sr_{0.15}MnO_3$ compound

Soma Das [a], D. Dhak [c], M.S. Reis [a], V.S. Amaral [a], T.K. Dey [b,*]

[a] Department of Physics, University of Aveiro, Portugal
[b] Cryogenic Engineering Centre, Indian Institute of Technology, Kharagpur, West Bengal 721302, India
[c] Material Engineering Department, University of British Columbia, Canada

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ABSTRACT

Polycrystalline $La_{0.7}Ba_{0.15}Sr_{0.15}MnO_3$ compound shows giant magnetoimpedance (GMI) effect at low field and at low frequency under a temperature range from 300 K to its Curie temperature (T_C). A sharp change in magnetoimpedance is obtained at low fields up to 1 kG, followed by a nearly linear increase at higher field. The frequency dependence of magnetoimpedance becomes prominent near T_C only, where as near room temperature; magnetoimpedance shows feeble frequency dependence in MHz region. Present investigation reveals that magnetoimpedance in $La_{0.7}Ba_{0.15}Sr_{0.15}MnO_3$ compound could be exploited as a sensitive tool for room-temperature magnetic field sensing.

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Magnetic materials with unusual magneto-transport properties take much interest in recent time because of their theoretical challenges and the potential applications in magnetic technologies. One such material is mixed-valence perovskite manganite that exhibits the colossal magnetoresistance (CMR) phenomena near their T_C. Apart from CMR, ac negative GMI effect, defined as $(\Delta Z/Z) = (Z(H) - Z(H=0))/(Z(H=0))$, has also been observed in these materials and is promising for developing highly sensitive magnetic field sensor [1–9]. In the present communication, we investigated the ac response of polycrystalline $La_{0.7}Ba_{0.15}Sr_{0.15}MnO_3$ and its temperature dependence from room temperature to its T_C over a frequency range from 50 kHz to 15 MHz and under a biasing dc magnetic field up to 4 kG. The maximum MI obtained at room temperature at 1 kG field at 50 kHz frequency is −66.4% and at 1 MHz is −72.6%. The result is interesting for use of this material as magnetic sensor applications.

Powdered $La_{0.7}Ba_{0.15}Sr_{0.15}MnO_3$ is prepared by chemical route as described in details in Ray and Dey [10]. AC susceptibility is measured by a susceptometer at 111 Hz and 0.10 G field and T_C was deduced from the minimum of $\delta\chi/\delta T$ vs. T curve. The approximate cylindrical-shaped sample is used for measurement of impedance. A signal coil of 25 turns was taken and the impedance of the coil with and without the sample is measured with an impedance analyzer (WK 6520A) at different temperatures. The biasing dc field up to 4 kOe is applied parallel to the exciting ac field. The resistive and reactive components of impedance are measured up to 15 MHz using excitation voltage of 0.5 V amplitude resulting in an ac field of ∼0.9 Oe.

XRD of this sample confirms the single phase nature and rhombohedral lattice structure (space group $R\bar{3}C$). Rietveld refinement gives the structural parameters as $a = b = 5.4977(0.0006)$ Å, $c = 13.5253(0.0021)$ Å. The tolerance factor t [$= (r_A + r_O)/(\sqrt{2}(r_B + r_O))$], r_A, r_B, r_O are ionic radii of A, B, oxygen for ABO_3 type compounds, comes out to be 0.865, which is higher than that for $La_{0.7}Sr_{0.3}MnO_3$ (0.853). This increase in tolerance factor due to Ba here signifies the release of structural strain in $La_{0.7}Ba_{0.15}Sr_{0.15}MnO_3$ [11,12]. However, the size-mismatch (variance) σ^2 of the A-site ionic radii, defined as $\sigma^2 = \sum y_i r_i^2 - \langle r_A \rangle^2$ [13], where r_i corresponds to various cation radii of A-site, y_i is their fractional occupancies and $\sum y_i = 1$, is increased also. This increased size mismatch does not seem to affect the material property much as observed by high Curie temperature ($T_C \sim 357.5$ K) of this sample very close to that of $La_{0.7}Sr_{0.3}MnO_3$ (∼370 K). This high T_C enables us to get well-defined metallic state at 300 K and a good magnetoimpedance effect is expected to occur accordingly. The magnetization isotherm (at 300 K) also gives characteristic ferromagnetic behavior and becomes saturated at ∼1.0 kG. The scanning electron microscopy of the present sample (Fig. 1) also gives a clear grain boundary with hexagonal-shaped particles (average particle size is 200 nm). The ac frequency dependence of the resistive (R) and reactive (X) part of the impedance (Z) is measured at different studied temperatures from room (300 K) up to 353 K (below T_C) and at different field from $H = 0$–4 kG and the impedance Z ($= \sqrt{R^2 + X^2}$) is obtained as shown in Fig. 2. A sharp increase in Z is obtained at frequency above a critical value (∼1 MHz) for all the studied temperatures. The value of this critical frequency corresponds to the

* Corresponding author.
E-mail address: tapasdey@hijli.iitkgp.ernet.in (T.K. Dey).

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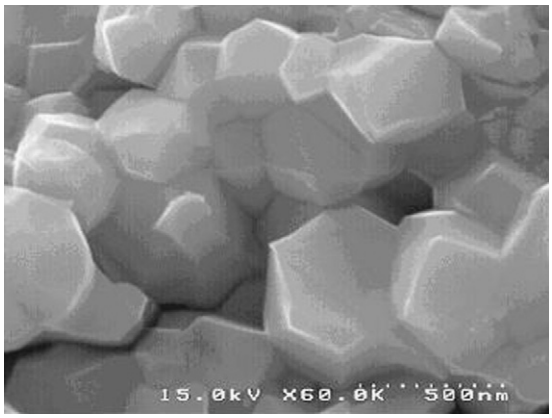

Fig. 1. Scanning electron micrograph of the studied $La_{0.7}Ba_{0.15}Sr_{0.15}MnO_3$ compound.

condition $d/\delta_m = 1$ where $2d$ and δ_m are the thickness of the sample and the penetration depth, respectively. For our sample, this penetration depth at this critical frequency comes out to be ~0.75 mm. A substantial decrease in Z with field is clearly obtained (Fig. 2) for all the temperature which is responsible to exhibit MI of the system. In general, MI effect is explained in terms of screening of electromagnetic field in magnetic metallic system according to classical electrodynamics [1,3,4,14]. With the increase of ac frequency (f), the penetration depth δ ($\delta = \sqrt{(2\rho/(\omega\mu_\varphi))}$; ρ is the resistivity, μ_φ is the transverse permeability for the circumferential field generated with ac current, $\omega = 2\pi f$) decreases and the charge carriers therefore prefer to flow near the surface of the material leading to high value of impedance with frequency. In case of soft ferromagnet, the high value of permeability μ_φ also decreases penetration depth δ and impedance increases correspondingly. Under a large dc magnetic field, because of nearly complete alignment of all the spins parallel to biasing field, the material becomes magnetically homogeneous and the permeability drops to low value giving high value of penetration depth and consequently large MI effect results. Fig. 3(a) and (b) shows the field dependence of R, X, Z at 300 K at 50 kHz and 15 MHz frequencies. As obtained, all the three quantities becomes field independent above 1 kG below which a sharp change is obtained at all the frequencies. Same behavior is obtained at all the temperature points studied between 300 and 353 K. Since in case of bulk polycrystalline manganites, grain boundary was proved to affect the transport and magnetic properties and hence the CMR properties significantly [15,16], the field insensitiveness of impedance above a critical field below T_C is expected to come from the complete spin alignment of the grain boundaries of this polycrystalline material. In the ferromagnetic metallic region of the polycrystalline manganites, the spins of the charge carriers

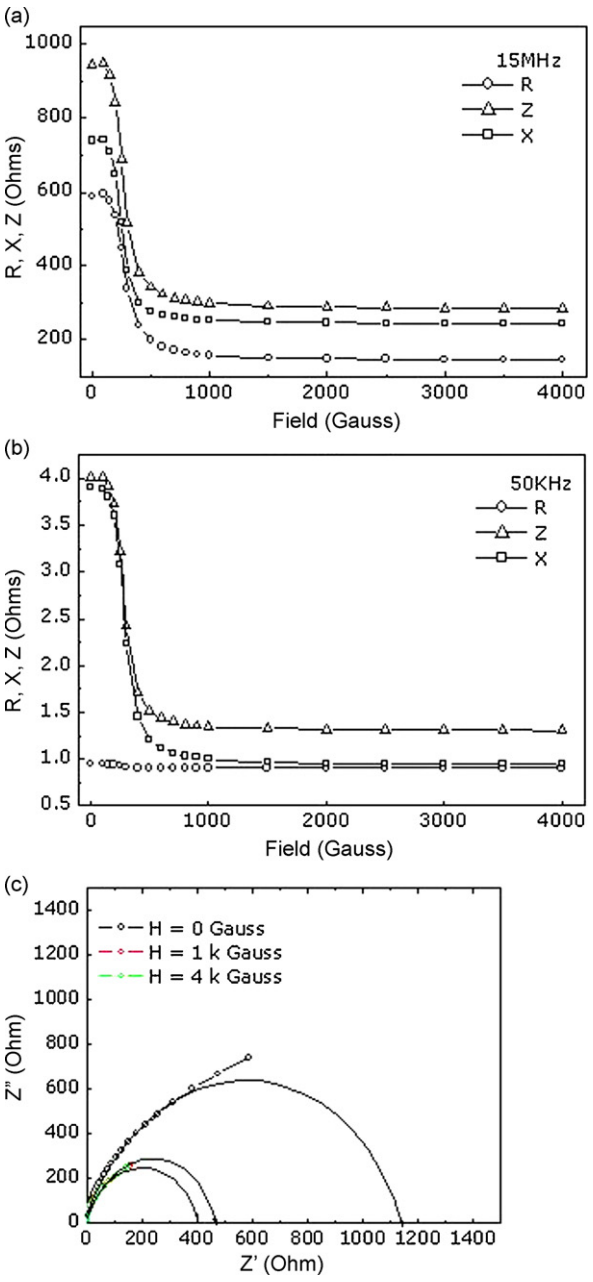

Fig. 3. (a and b) Typical room temperature field dependence of R, X and Z at 50 kHz and 15 MHz frequencies. (c) Cole–cole plot for $La_{0.7}Ba_{0.15}Sr_{0.15}MnO_3$ compound at room temperature.

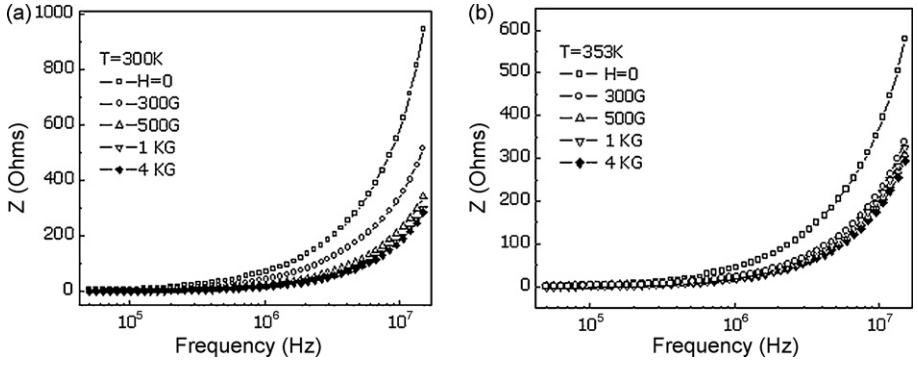

Fig. 2. Frequency dependence of impedance (Z) of $La_{0.7}Ba_{0.15}Sr_{0.15}MnO_3$ compound at different fields at temperatures $T = 300$ and 353 K.

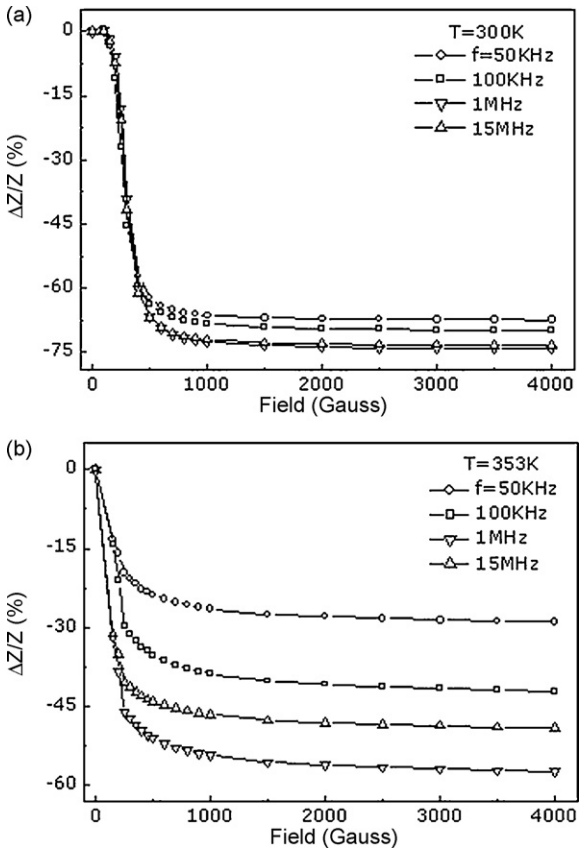

Fig. 4. Field dependence of $\Delta Z/Z$ with magnetic field at four different frequencies for $T = 300$ and 353 K.

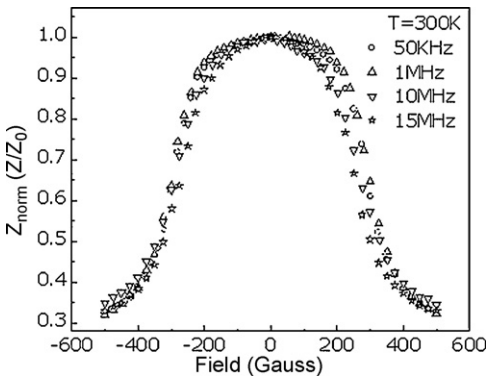

Fig. 5. Field dependence of Z/Z_0 at ±500 G at room temperature for four different frequency ranges and linear variation of $\Delta Z/Z$ between ±200 and ±350 G magnetic field for all frequencies covered.

are aligned within the grains where as a magnetically disordered region is obtained near the grain boundaries. The spins are canted in this grain boundary region with a distribution of angles and are aligned with increasing magnetic field. This agrees with the fact that the magnetization continues to increase with field in the polycrystalline samples [15]. On application of a high magnetic field, the increasing alignment of the neighboring spins in the disordered region leads to a reduction in resistance as well as reactance and hence the impedance. The field independent behavior of Z of this system above 1 kG originates due to complete spin alignment across grain boundaries, which is also reflected on the behavior of magnetization isotherm, where magnetization gets almost saturated at ~1 kG. Further, the complex plot between the real and imaginary components of the impedance in the studied frequency range is plotted in Fig. 3(c). It may be seen that the dc conductivity obtained from Fig. 3(c) increases from 0.0109251 Ω^{-1} cm^{-1} at zero field to 0.0307518 Ω^{-1} cm^{-1} at 4 kG. Typically, manganites in ferromagnetic metallic state have the resistivity in milliohm-cm range [19], the observed high value of resistivity (cole–cole plot) may therefore be assumed to originate from more resistive grain boundaries present in the material.

The dc magnetic field dependence of MI (=$\Delta Z/Z$) of this system at different temperatures and frequencies is shown in Fig. 4. An increase in MI value with increasing frequency up to 1 MHz is obtained giving a maximum value of ~74% and it drops marginally to ~70% at 15 MHz at room temperature. This decrease in MI values at higher frequencies is obtained since the excitation field at higher frequencies flows through a very thin layer near the surface and the sample becomes insensitive to the ac field. Similar behavior in MI is obtained for other temperatures also. However, decrease of MI of this system with increasing temperature arises from the increasing magnetic inhomogeneity introduced in to the system with increasing influence of paramagnetic component. The permeability of the system decreases with increase of paramagnetism, the penetration depth increases, consequently Z decreases with temperature. Under the application of an external magnetic field, the spin alignment occurs across the grain boundaries depending on the frequency which influences the permeability and we therefore get a prominent frequency dependence on MI at high temperature near its T_C (353 K) in Fig. 4(b). Frequency therefore plays a vital role in controlling permeability at high temperature, which is reflected in the change in impedance Z. Another important feature is that the field dependence of MI observed in this system exhibits the striking similarity with the field dependence of dc magnetoresistance observed in granular manganites [17–19]. The sharp change in dc magnetoresistance at lower field followed by a nearly linear decrease at higher field in granular manganites is already proved to be due to spin polarized tunneling of the charge carriers across the grain boundaries [17]. This similarity in behavior indicates major role of grain boundaries behind the sharp fall in MI observed at lower fields.

Fig. 5 shows the normalized Z value ($Z_{norm} = Z/Z_{H=0}$) at ±500 G at room temperature at four different frequencies. Strikingly, the sharp change in Z_{norm} is linear in a restricted field zone of ~200–50 G, where MI gives a linear increase from ~7 to 61%. This behavior of MI appears to be related to the granular nature of the sample. The field (~200 G) at which MI display a sharp change, correspond to the minimum field needed to start the spin alignment of the charge carriers across the grain boundaries in this polycrystalline material. However, the change in MI is very nominal and is between 6 and 8% at all frequencies. This small change observed at fields below 200 G is expected to be due to the domain wall motion inside the grains of this manganite material; however it needs further support to conclude about domain wall bulging on application of external magnetic field.

In conclusion, we have presented the frequency and field dependences of the magnetoimpedance in $La_{0.7}Ba_{0.15}Sr_{0.15}MnO_3$ compound. Both resistive and reactive components of impedance increase with increasing ac frequency due to skin effect of the metallic state below T_C. Above a critical frequency value of 1 MHz, a sharp increase in both R and X and hence Z is obtained for this system. A large change in magnetoimpedance (>70%) has been observed only at low fields up to 1 kG, beyond which MI nearly saturates. The observed behavior is explained in light of the dependency of skin depth on external magnetic field and the applied frequency. We further suggest a significant role of spins across the grain boundaries and their influence under applied magnetic fields. The present study also indicates that giant magnetoimpedance (>70%) observed at room temperature in $La_{0.7}Ba_{0.15}Sr_{0.15}MnO_3$ compound could

be exploited as sensitive and inexpensive magnetic sensor.

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