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Size dependence in magnetic memory, relaxation and interaction of $La_{0.67}Sr_{0.33}MnO_3$

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ABSTRACT

We report the study of memory effect, relaxation and interaction of $La_{0.67}Sr_{0.33}MnO_3$ (LSMO) particles of two different sizes, bulk (~60–150 μm) and nano (~80 nm) synthesized by using polymeric precursor route. Magnetic relaxation measurement has been carried out and the analysis shows the existence of two relaxation times in both the cases. Our dynamic light scattering (DLS) studies indicate the presence of two different size distributions in LSMO nano particle sample. This is attributed to the fact that particles of two different sizes can relax separately. It has been observed that both the samples show signature of magnetic memory effect. Because, the effect of disordered surface layer is more in LSMO nano than LSMO bulk particles, LSMO nano particles are found to favor demagnetized state where as LSMO bulk prefer magnetized state.

1. Introduction

Observation of magnetic memory in magnetic nano particles is an interesting phenomenon, since the reason can be either attributed to interparticle interaction or particle size distribution. This is a potential area for recent scientific activities because of the enormous technological application and exotic physics governed by different competing effects. Most of such works have been focused on ferromagnetic and ferrimagnetic nanoparticles due to their high magnetic moments which make them industrially valuable. Hole doped perovskite manganites are well known magnetic materials for their fascinating physical properties and technological viability. However, the effect of size reduction on their physical properties is a topic of very recent interest [1–10]. In general, properties of nanoparticles of magnetic materials can arise from intrinsic properties, or from factors arising from interactions of nanoparticles [11,12]. Depending on the composition and hole-doping level, many manganites exhibit unusual nonequilibrium dynamics and time-dependent phenomena such as ageing, rejuvenation, memory, etc. [13–16]. $La_{0.67}Sr_{0.33}MnO_3$ (LSMO) belongs to hole doped manganite family and is known to be potential candidate for technological application, since it has ferromagnetic transition temperature (T_C) around 380 K and a large magnetic moment at room temperature [17–21]. The present work is concentrated on the study of memory effect and relaxation phenomena of LSMO bulk and nano particles of various sizes. We report on the results of our detailed investigation in the non-equilibrium magnetic behavior of LSMO nano and bulk. The main objective is to understand the type of interaction and to investigate the effect of size reduction on magnetic interactions, since interaction plays important role to influence the magnetic properties.

2. Experimental details

The LSMO nano particles have been synthesized by the sol–gel based polymeric precursor route. In a typical synthesis process, precursor materials are dissolved in the desired stoichiometric proportions in appropriate solvent and polymer (Ethylene Glycol). The mixture has been heated till the sol is formed. The gel has to be dried. Pyrolysis is to be done to remove organic precursor materials around 300–400 °C followed by sintering around ~650 °C in order to obtain the desired chemical phase [22]. All the synthesized nano particles have been characterized using powder X-Ray Diffraction (XRD) using CuKα radiation at room temperature. The particle size distribution is determined by dynamic light scattering (DLS). The temperature at which the DLS has been done is 25 °C and the solution is 2-propanol where the nano particles have been dispersed. LSMO nano particles are taken in required quantity and pellets are made at high pressure (around 100 MPa). The pellets are heated at 1100 °C for 10 h and 1400 °C for 12 h

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which yields LSMO bulk particles. These particles are characterized by Scanning Electron Microscopy (SEM). We have carried out Magnetization (M) vs. Magnetic field (H) measurements at 300 K, 85 K and 120 K by Vibrating Sample Magnetometer (VSM). Zero field cooled (ZFC) and field cooled (FC) magnetization measurements at $H=200$ Oe have shown appreciable difference. For ZFC measurements at $H=40$ Oe with 2 h stop at 90 K when compared with the uninterrupted ZFC measurements, we found the signature of magnetic memory effect. In order to do magnetic relaxation measurement, the sample is cooled down to 95 K in the absence of any field. Then a field $H=1$ T is applied in 20 s and it took again around 20 s to remove the field. After removing the field, the magnetization is recorded as a function of time ($t=7$ h). In order to probe the inter-particle interaction for the LSMO nano and bulk samples, the measurement of isothermal remnant magnetization (m_{IRM}) and dc demagnetization (m_{DCD}) as a function of magnetic fields have been carried out [23]. We have investigated the validity of Wohlfarth relation in the LSMO particles; which is applicable for a non-interacting system with uniaxial anisotropy [24].

3. Results

The XRD data shown in Fig. 1 determines the required phase formation for LSMO nano particles. The positions of intensity peaks are compared and observed to be consistent with the data base of International Centre for Diffraction Data (ICDD). The results of DLS experiment of nano LSMO dispersed in 2-propanol are described in Fig. 2. The DLS data have been fitted with gaussian function and the error bar is around 1%. The DLS data show 2 peaks, – one (around 80 nm) for nano particles and other (around 185 nm) due to particle agglomeration. It seems that there are particles of 2 different sizes existing (one real size of nanoparticle and another is agglomerated size), – very small and very large. The average particle size is around 80 nm as it is determined by DLS data analysis. The LSMO bulk particles have been studied by SEM and the results are reproduced in Fig. 3. It is seen that there are particles with the size range of 60–170 μm.

The initial M vs. H measurements for LSMO nano particles have been carried out at 85 K and 300 K. The curves show clear hysteresis which suggests the existence of qualitative ferromagnetic phase for LSMO nano at 85 K and 300 K. Although the complete saturation has not been achieved at 10,000 Oe applied field as shown in Fig. 4(a) and (b). Similar measurement has been done for LSMO bulk at 300 K which also shows hysteresis and better

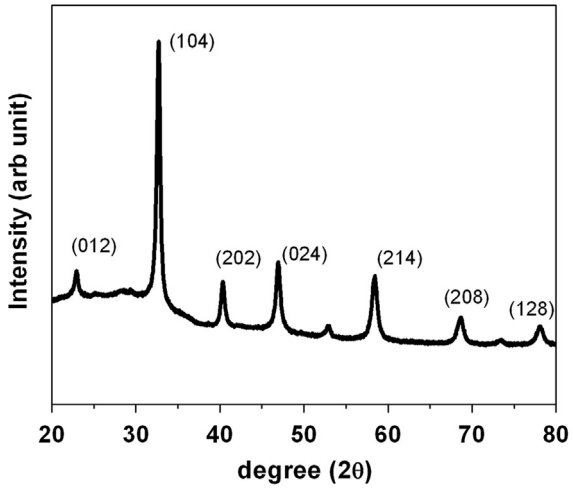

Fig. 1. XRD pattern for LSMO nano where the line is experimental data.

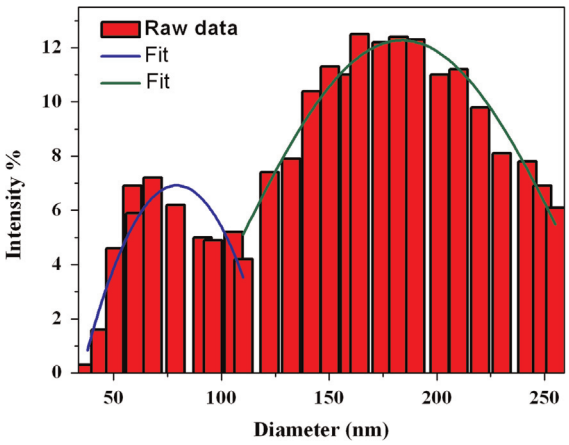

Fig. 2. Results of dynamic ligh scattering (DLS) experiment of nano LSMO dispersed in 2 propanol showing 2 peaks, – one for nano particles and other due to particle agglomeration. The bar symbols are raw data and solid lines are gaussian fits.

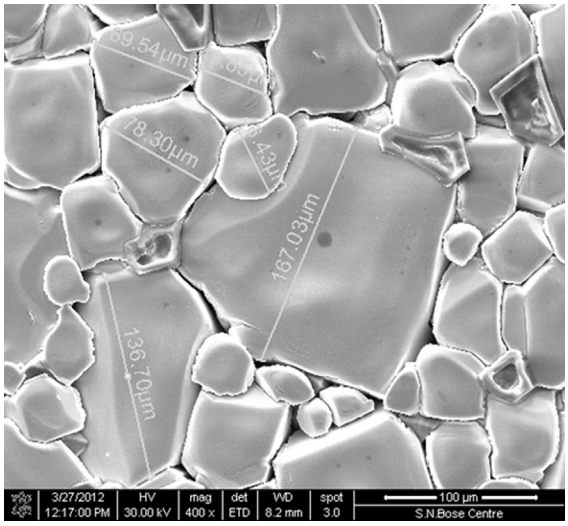

Fig. 3. Results of SEM study of bulk LSMO showing particles with the size range of 60–170 μm.

saturation at 10,000 Oe in comparison to LSMO nano (see Fig. 4 (c)). We have measured the magnetization of LSMO nano with respect to the temperature; while warming after cooling the sample in absence (ZFC) and in the presence (FC) of $H=200$ Oe magnetic field. This sample shows bifurcation between the ZFC and FC profiles (Fig. 4(d)) which is a typical behavior for LSMO nano particles [25]. There is a peak in ZFC curve and FC magnetization shows an irreversible behavior with respect to that of ZFC below the peak temperature. This indicates that below the peak temperature the magnetic system goes onto a disordered blocked state. The blocking temperature T_B has been determined as around 195 K from the peak of the ZFC curve (see Fig. 4(d)). It is to be noted that the obtained T_B is the average of the T_B's within the sample because of finite size distribution of the particles. The temperature at which the bifurcation starts between ZFC and FC is known as the irreversible temperature (T_{IR}) and it is found to be 270 K.

We have cooled the sample down to 90 K at zero field and waited for 2 h at 90 K in zero field. Then we have further cooled the sample down to 80 K. At 80 K, we have applied the field of 40 Oe and taken the ZFC data at heating cycle. We have again cooled the sample down to 80 K at zero field. Then at 80 K we have applied the field of 40 Oe and taken the ZFC data at heating cycle