

# Magnetism of Nanoparticle $\text{La}_{0.7}\text{Ca}_{0.3}\text{Mn}_{0.7}\text{Fe}_{0.3}\text{O}_3$ under Applied Hydrostatic Pressure

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Nanoparticle  $\text{La}_{0.7}\text{Ca}_{0.3}\text{Mn}_{0.7}\text{Fe}_{0.3}\text{O}_3$  with the average particle size of 10 nm was mechanochemically synthesized in the single-step procedure. Temperature and field dependences of magnetization were recorded under both ambient and applied hydrostatic pressure up to 0.47 GPa. At ambient pressure, two magnetic transition points were observed: one of the spin-glass like type at the temperature  $T_f \approx 34$  K, and ferro-to-paramagnet transition at  $T_C \approx 51$  K. Under the applied pressure magnetic parameters gradually changed: (i) both  $T_C$  and  $T_f$  were lowered with the increase in pressure; (ii) both high field magnetization  $M_{5T}$  and remanent magnetization  $M_{\text{Rem}}$  decreased with pressure; (iii) coercivity  $H_C$  increased with pressure. The obtained results show that magnetism of a nanoparticle system with high degree of intrinsic disorder diminishes with the applied pressure that is in compliance with the effect of the increased cationic disorder (“internal pressure”) in bulk manganites.

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## 1. Introduction

Perovskite manganites with chemical formula  $\text{A}_{1-x}\text{A}'_x\text{MnO}_3$  (A = rare earth, A' = alkali earth cation) have been extensively investigated, because they possess unique combination of a variety of physical properties. Typical mixed valent manganite representative and intensively studied compound  $\text{La}_{1-x}\text{Ca}_x\text{MnO}_3$  possesses rich phase diagram where transitions can be influenced by chemical doping, applied magnetic field and hydrostatic pressure [1, 2]. Partial substitution of iron for manganese introduces additional perturbations that can lead, in combination with the reduction of particle size down to nanometric scale, to emergence of new physical properties. Investigation of pressure effects on such a system can provide valuable information on the underlying physical mechanisms responsible for these new properties.

## 2. Experimental

Nanoparticle  $\text{La}_{0.7}\text{Ca}_{0.3}\text{Mn}_{0.7}\text{Fe}_{0.3}\text{O}_3$  was obtained by mechanochemical milling of corresponding metal-oxide powders by using planetary micro mill Fritsch Pulverisette 7 with vials and balls of tungsten carbide, in the ambient conditions. The synthesis process was controlled by the X-ray diffraction (XRD) at each half hour

of the milling time. After 4.5 h single-phase perovskite compound of the orthorhombic space group  $Pnma$  (ICSD 155409) was obtained with no noticeable changes in recorded patterns for prolonged milling time. The average particle size was determined to be 10 nm by high resolution transmission electron microscope (HRTEM), Fig. 1. Magnetic measurements under applied hydrostatic pressure were performed on the SQUID-based magnetometer MPMS XL-5 by utilising specialized CuBe pressure cell filled with the mixture of mineral oils [3].

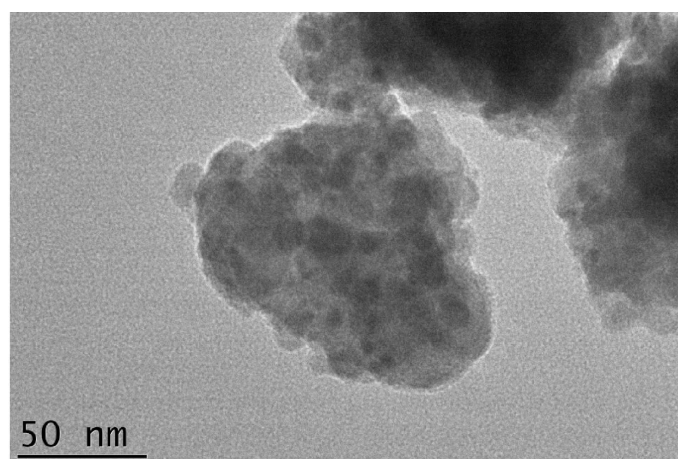


Fig. 1. HRTEM micrograph of investigated  $\text{La}_{0.7}\text{Ca}_{0.3}\text{Mn}_{0.7}\text{Fe}_{0.3}\text{O}_3$  sample.

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### 3. Results

Temperature dependence of magnetization measured at ambient pressure reveals existence of two magnetic transitions (Fig. 2): of the spin-glass type at  $T_f \approx 34$  K (additionally verified by AC-susceptibility measurements, not shown here), as well as ferro-to-paramagnet transition at  $T_C \approx 51$  K (both critical temperatures were determined from  $dM/dt$  dependence). Isothermal magnetization vs. field data confirms frustrated nature of low-temperature magnetic state since it is combined of hysteretic behaviour at low fields and almost linear non-saturated high field part (Fig. 3). The same magnetic

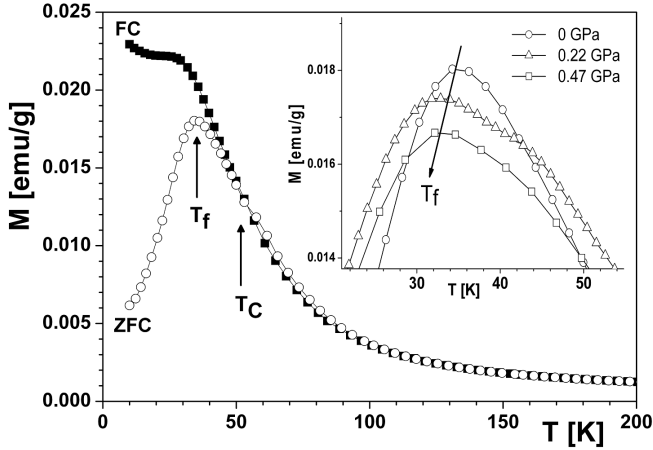


Fig. 2.  $M(T)$  dependence at ambient pressure in ZFC and FC regimes. Inset: shift of  $T_f$  with applied pressure.

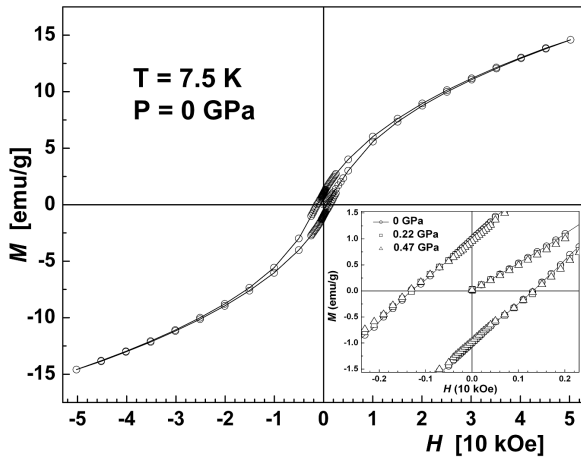


Fig. 3. Magnetic hysteresis at ambient pressure. Inset: change of magnetization data with applied pressure.

measurements were repeated under the applied pressure of 0.22 GPa and 0.47 GPa, and following changes of magnetic parameters were noticed: (i) both transition temperatures  $T_f$  and  $T_C$  decrease with the pressure increase;

(ii) both remanent magnetization  $M_{Rem}$  and magnetization at highest field applied  $M_{5T}$  decrease with pressure; (iii) coercivity  $H_C$  increases with pressure. Obtained values of magnetic parameters are listed in Table.

TABLE

Selected magnetic parameters at ambient and two applied pressures.

P [GPa]	$T_C$ [K]	$T_f$ [K]	$M_{5T}$ [emu/g]	$M_{Rem}$ [emu/g]	$H_C$ [Oe]
0	51.2	33.5	14.6	1.0	1310
0.22	50.4	32.8	14.3	0.98	1330
0.47	49.3	32.0	14.0	0.96	1340

### 4. Conclusion

Mixed valent manganite  $La_{1-x}Ca_xMnO_3$  is well-known as a system with the presence of both ferro- and antiferromagnetic interactions. Both nanosizing and partial substitution of iron for manganese introduce additional magnetic disorder and/or frustration, and so the investigated nanoparticle  $La_{0.7}Ca_{0.3}Mn_{0.7}Fe_{0.3}O_3$  possesses re-entrant spin-glass-like behaviour that speaks in favour of high degree of intrinsic magnetic frustration. Our results show that application of external pressure on such a system further increased internal disorder since magnetism of the system diminishes with the pressure increase. This result is in compliance with the conclusions drawn for nanoparticle  $La_{0.7}Ca_{0.3}MnO_3$  with high degree of structural disorder [4] as well as with the effects of increased cationic disorder (i.e. “internal pressure”) in bulk manganites [5].

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### References

- [1] E. Dagotto, T. Hotta, A. Moreo, *Phys. Rep.* **344**, 1 (2001).
- [2] V. Markovich, I. Fita, D. Mogilyansky, *J. Phys., Condens. Matter* **19**, 346210 (2007).
- [3] J. Kamarad, Z. Machatova, Z. Arnold, *Rev. Sci. Instrum.* **75**, 5022 (2004).
- [4] V. Kusigerski, D. Markovic, V. Spasojevic, M. Tadic, M. Zentkova, M. Mihalik, *J. Nanopart. Res.* **12**, 1299 (2010).
- [5] L.M. Rodriguez-Martinez, J.P. Attfield, *Phys. Rev. B* **54**, R15622 (1996).