

Spin Glass Dynamics of Nanoparticle $\text{La}_{0.7}\text{Ca}_{0.3}\text{Mn}_{0.7}\text{Fe}_{0.3}\text{O}_3$ Obtained by a Mechanochemical Milling

M. PEROVIC*, A. MRAKOVIC, V. KUSIGERSKI, J. BLANUSA AND V. SPASOJEVIC

Condensed Matter Physics Laboratory, Vinca Institute, University of Belgrade, P.O.B. 522, 11001 Belgrade, Serbia

Spin dynamics of nanoparticle $\text{La}_{0.7}\text{Ca}_{0.3}\text{Mn}_{0.7}\text{Fe}_{0.3}\text{O}_3$ system was studied through the set of diverse magnetic measurements. Analysis of the data obtained from magnetic relaxation measurements, memory effect and AC susceptibility experiments pointed to a spin glass like behavior of interacting nanoparticle system.

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1. Introduction

Current efforts in nanoparticle magnetism research are in large part dedicated to the investigations of the magnetic properties of interacting systems [1]. Among most attractive topics are slow dynamics, aging and memory effects. In this paper we investigate the above effects on the nanoparticle mixed valence manganite $\text{La}_{0.7}\text{Ca}_{0.3}\text{Mn}_{0.7}\text{Fe}_{0.3}\text{O}_3$, additionally frustrated by iron doping at Mn-crystallographic site. Magnetic measurements (AC susceptibility, zero-field cooled (ZFC) DC magnetization, and relaxation measurements) are performed on the Quantum Design MPMS XL-5 SQUID magnetometer.

2. Results and discussion

AC susceptibility measurements have been performed in the temperature range 10–60 K at four frequencies ν with the drive amplitude of AC field 4 Oe, Fig. 1. Frequency dependent cusp was observed in both in phase χ' and out of phase χ'' components. Evidence of spin glass like behaviour was obtained from the analysis of the frequency dependence of T_{\max} . Relaxation time τ in spin glass systems diverges as a power law $\tau = \tau_0[(T_{\max}/T_G) - 1]^{-z\nu}$ (Fig. 1, inset), where τ_0 represents the shortest relaxation time available to the system, $z\nu$ is dynamical critical exponent, and T_G temperature is determined by interactions in the system. Parameter value $z\nu = 7.6$ obtained from the fit is expected for spin glass systems (7–14), although $\tau_0 = 1.6 \times 10^{-10}$ s is higher than typical values for spin glasses ($< 10^{-11}$ s). Temperature T_G was 34.8 K.

Spin-glass like behaviour is also manifested by a memory effect in ZFC DC magnetization, which was measured following the procedure described in [1]. During

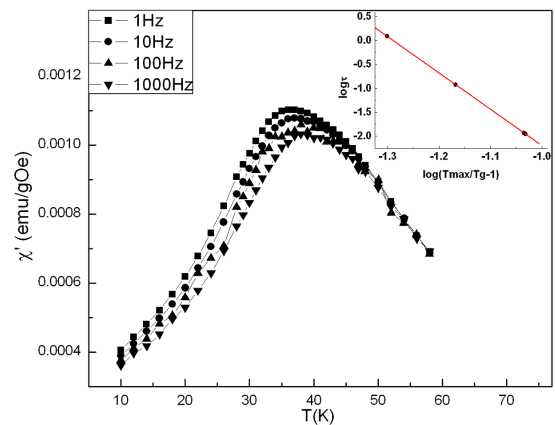


Fig. 1. Temperature dependence of the real part of the AC susceptibility.

the procedure, cooling was stopped at $T_W = 20$ K for the time t_W . $M(T)$ curves were recorded for waiting times $t_W = 1, 3, 7$ and 10 h, Fig. 2. Clear difference between measured curves and the reference curve can be noticed. The $M(T) - M_{\text{ref}}(T)$ difference shows a pronounced dip around waiting temperature $T_W = 20$ K (Fig. 2, inset). Above T_{SGL} differences between the curves vanish, showing that memory effect below T_{SGL} is assigned to a spin glass state. ZFC memory effect is a consequence of spin correlation and is not found in superparamagnetic systems.

The aim of the present study was to investigate the influence of the interparticle interactions on relaxation processes. As the interactions become appreciable, relaxation processes become correlated. Interparticle interactions modify energy barriers, making energy of the system a multivalley structure, like that in spin glasses.

Theoretical approach of hierarchically constrained dynamics, proposed by Palmer et al. [2], treats correlated

* corresponding author; e-mail: mara.perovic@vinca.rs

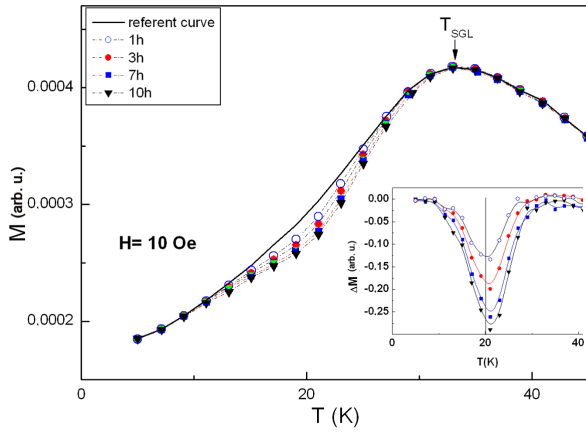


Fig. 2. ZFC memory effect for different waiting time.

relaxation processes. Although it was originally proposed for description of slow collective dynamics in spin glass systems, it is also adequately used in the case of interacting fine particles [3]. A distribution of relaxation times, which is the origin of the slow relaxation, arises from the volume anisotropy energies distribution and interactions among the particles. Magnetization relaxation in interacting systems follows the stretched exponential form. $M(t) = M_0 + M_r e^{-(t/\tau)^{1-n}}$, where M_0 is related to an intrinsic ferromagnetic like component, and M_r to a glassy component contributing to the relaxation effect. The time constant τ and the parameter n are related to the relaxation rate S , defined as $S = -\frac{1}{m_0} \left(\frac{dm}{d \ln t} \right) = f(E^*) k_B T$, which mirrors energy barriers distribution $f(E^*)$ [4].

Magnetic relaxation curves were measured in the following way: sample was cooled to the measuring temperature T_m in the field 300 Oe. After the waiting time t_W , field was switched off and time dependence of isothermal magnetization was measured for 10^4 s. Magnetization relaxation was measured for $t_W = 0$ s and $t_W = 3600$ s. In order to investigate aging process and obtain relaxation time spectrum, ZFC magnetization measurements were performed using the same procedure, with cooling in zero field and application of weak field of 2 Oe after t_W . Between measurements, the sample was reheated to 200 K, well above blocking temperature $T_B = 51$ K (not shown here).

Experimentally obtained FC and ZFC magnetization curves were accurately characterized by stretched exponential, Fig. 3. Magnetization curves were sensitive to the magnetothermal history, with magnetization values changing with t_W . Time variations of derived relaxation rate S at temperature $T_m = 20$ K for ZFC case is plotted in Fig. 4. At all temperatures T_m , pronounced t_W dependence of the magnetization and relaxation rate S curves is observed. Relaxation rate S shows maximum at the observation time close to t_W , which corresponds to an inflection point on $M(t)$ plot.

These features were observed in the variety of experiments on spin glass systems. Aging phenomena in

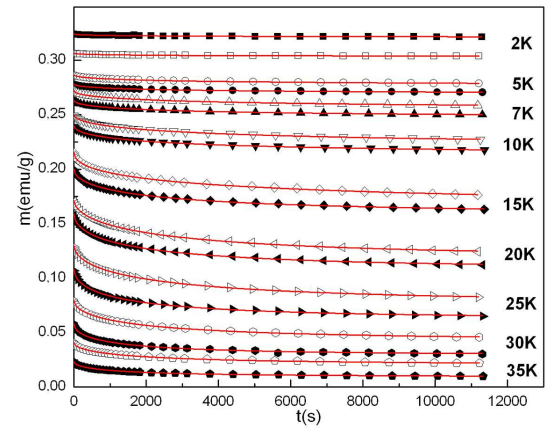


Fig. 3. Relaxation FC magnetization curves at temperatures T_m , $t_W = 0$ s (full symbols) and $t_W = 3600$ s (open symbols).

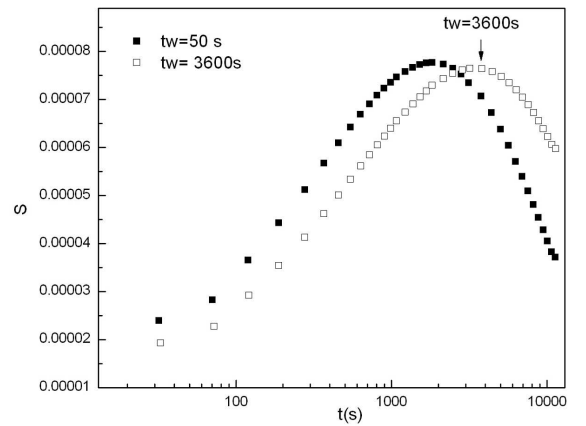


Fig. 4. Relaxation rate S derived from ZFC magnetization curve at 20 K.

$\text{La}_{0.7}\text{Ca}_{0.3}\text{Mn}_{0.7}\text{Fe}_{0.3}\text{O}_3$ represent a clear indication of strong interactions with a phenomenology which mimics that of spin glasses.

Acknowledgments

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