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Size distribution effect on the magnetization and magneto-resistance in ferromagnetic nanostructured manganites

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Abstract

We present experimental data of magnetization and magneto-resistance of nanostructured $La_{2/3}B_{1/3}MnO_3$ with B = Ca, Sr, which present difference between the coercive field in the magnetization loop with their corresponding maximum value in the magneto-resistance. This difference is described by a model that include, size distribution of magnetic particles, randomly oriented anisotropy axis and electronic transfer between the particles, which is mediated by spin-polarized tunneling process. Also, the model predicts that the maximum magneto-resistance can be, in the magnetic disorder state, two times larger than the experimental value. The model results can be used to estimate the size dispersion of nanoparticles in similar systems.

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1. Introduction

The transport and magnetic properties of the $La_{0.67}B_{0.33}MnO_3$ (B = Ca, Sr, Ba) compounds have been considerably studied after the discovery of the 'colossal magneto-resistance' (CMR). The metallic and ferromagnetic character, below T_C, was early described in terms of the double exchange (DE) mechanism [1–3]. In single crystals and epitaxial films, the magnetic field dependence of the magneto-resistance (MR) decreases linearly from zero. However, polycrystals and granular materials show a rapid decrease of MR called 'low field magneto-resistance' (LFMR) [4,5], which is an 'extrinsic' effect. This effect was also observed in non-multilayer system with giant magneto-resistance using immiscible alloys with non-aligned ferromagnetic entities immersed in a metallic media (Co–Cu; Fe–Cu) [6]. On the other hand, this MR drop has been also seen in cold-pressed powders of half-metallic CrO_2 ferromagnet, in dilution of these particles in anti-ferromagnetic Cr_2O_3 [7], in polycrystalline thin film and in powder compact of the Fe_3O_4 spinel [8]. The LFMR has generated a great interest since it is present in a wide temperature range, making granular materials good candidates for potential applications [9].

The LFMR has been intensively investigated [5,10–14]. Some authors [4,15] have described it by the spin-polarized tunneling (SPT) of carriers across grain boundaries (gb) using the earlier SPT model introduced by Helman and Abeles (H–A) for granular Ni films [16]. In a previous paper, we have presented experimental data on ferromagnetic nanostructured $La_{0.67}Sr_{0.33}MnO_3$ [5] showing that H–A does not describe the experimental situation of granular manganites and in addition, we noted a marked difference between the field at which the magnetization is zero in the hysteresis loops and the maximum (minimum) of the magneto-resistance (magneto-conductivity) curve. To explain the LFMR of manganites, more realistic models

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for half-metallic materials have been proposed [17–19]. In particular, Sun and Li [20] discussed the temperature (T) dependence of MR in terms of electron jumps between superparamagnetic (SP) grains. On the other hand, a study of the field effect in SP grains of Cu–Co melt spun ribbons shows that H–A expected behavior is not observed and the results were explained including in a model inter-grain scattering and grain size distributions [21,22].

In the present work, for nanostructured Sr and Ca-doped manganites, we measured at low T the magnetization (M) and the magneto-conductivity (M_σ) simultaneously. Our experimental results can be understood with a simple model that include the essential ingredients for the magnetic and electrical transport properties in granular manganites, the size distribution of the particles and SPT transfer process. We also obtained that the model does not present an unequivocal relation between LFMR and M, and also it explains the experimental difference between the 'magnetic coercive field', H_c (the field where the magnetization is zero in the magnetization loop), and the 'magneto-conductive coercivity', H_σ (the field where the magneto-conductivity is minimum). The ratio H_c/H_σ can be used to estimate the grain size dispersion in a granular manganite.

2. The model

We simplify the problem performing simulations on a square lattice of $n^2=N$ sites (Fig. 1(b)). In each site is placed a grain of a given size being it a single magnetic domain (SMD) with magnetic moments blocked in the anisotropy direction and a H_c should be observed. The model also includes SPT conditions to reproduce the jump of the carriers between two adjacent grains. From the simulations M and M_σ data can be obtained at each H by the application of a difference of voltage between the electrodes (the ends of the square lattice used). In these 2D lattices realistic effects as non-homogeneous current

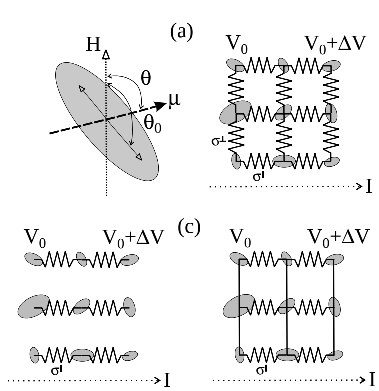

Fig. 1. (a) Magnetic field H, grain anisotropy axis (thin line) and grain magnetization μ. (b) General grain and conductances network. (c) Conductance network in the case of $\sigma^{\perp}=0$. (d) Conductance network in the case of $\sigma^{\perp}=\infty$.

and percolation paths can be observed. Our 2D calculus is between a 1D chain and the homogeneous case (mean field or infinite dimensional), while the 3D lattice behavior is expected between the mean field case and our calculus.

In the simulation, we considered that each grain has a characteristic size r, Zeeman energy $E_Z=-\mu(r)H\cos(\theta)$ and uniaxial anisotropy contribution $E_A=A(r)sin^2(\theta-\theta_0)$. Dipolar interaction between grains are neglected. The angles θ and θ_0 are determined by the M and the anisotropy axis with respect to H, respectively (Fig. 1(a)). For simplicity, E_Z, E_A and the conductivity (σ) are written in 2D angular expressions although in the calculus 3D forms are used. Each direction of grain anisotropy axis is defined randomly from an isotropic distribution. The magnetic moment ($\mu(r_i)$) of the ith grain is proportional to the grain volume $m(r_i)\propto r_i^3$. The magnetization is evaluated simply by adding each grain contribution

$$M(H)=\frac{1}{\sum_{i=1}^{N}\mu(r_i)}\sum_{i=1}^{N}\mu(r_i)\cos\theta_i \tag{1}$$

For the electric properties, we assumed two conductivity channels between neighboring grains. One of them (σ_{NM}) takes into account the flow of unpolarized electrons and it is independent of the magnetization [19]. This contribution may be due to surface, inter-grain contributions [18] or small defects inside of each grain, which induce to lose the spin polarization. Furthermore, this term is necessary to fit precisely the experimental data. Other necessary ingredients to be taken into account is the tunneling probability of transfer particles with spin $s=1/2$ between grains. This magnetic contribution to the conductivity is dependent of half the angle between the directions of each grain magnetization ($\Delta\theta=\theta-\theta'$). Specifically, the conductivity of the second channel can be described by the spin polarized tunneling [17], which takes the form $\sigma_M\cos^2(\Delta\theta/2)$ (we neglect the phase that appears for 3D classical spins [23]). Even though the experimental data showed that the transfer occurs in the Coulomb blockade regime, we assumed that the angular dependence of the tunneling probability takes the same form as that discussed by Raychauduri et al. and Dai et al. [17,18].

In the calculus of each M and σ value, we averaged approximately 30 runs over a 10^2 sites of the square lattice. We set the first column of the plane at a constant potential V_0 and the last column at a potential $V_0+\Delta V$ (scheme in Fig. 1(b)). With these boundary conditions, we calculate the angle for each grain in the presence of the external field. To obtain the current, we solved the set of Kirchoff's equations.

The inclusion of the grain size distribution, where each grain of size r has associated a magnetization module $|\mu(r)|$ and an anisotropy energy $A(r)$, leads to a distribution of H_c in the magnetization response. On the other hand, the effect on σ is not as easily and directly observable.