Eur. Phys. J. B **50**, 99–103 (2006)
DOI: 10.1140/epjb/e2006-00065-5

THE EUROPEAN PHYSICAL JOURNAL B

Physical meaning and measurement of the entropic parameter q in an inhomogeneous magnetic systems

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Received 19 October 2005 / Received in final form 5 December 2005
Published online 20 February 2006 –

Abstract. In this paper we present a thorough analysis of two systems magnetically inhomogeneous: the manganite $La_{0.7}Sr_{0.3}MnO_3$/MgO and the amorphous alloy $Cu_{90}Co_{10}$. In both cases, the non-extensive statistics yield a faithful description of the magnetic behavior of the systems. In the model proposed here, the inhomogeneous magnetic system is composed by many Maxwell-Boltzmann homogeneous bits and the entropic parameter q is related to the moments of the distribution of the inhomogeneous quantity. From the analysis of Scanning Tunnelling Spectroscopy (STS) images, the q parameter can be directly measured.

PACS. 05.70.Ce Thermodynamic functions and equations of state – 05.70.Fh Phase transitions: general studies

1 Introduction

Tsallis thermostatistics has been widely used in a number of different contexts [1]. The framework is applicable to systems which, broadly speaking, present at least one of the following properties: (i) long-range interactions, (ii) long-time memory, (iii) fractality and (iv) intrinsic inhomogeneity [1,2]. Manganese oxides, or simply manganites, seems to embody three out of these four ingredients: they present Coulomb long-range interactions [3–5], clusters with fractal shapes [6,7] and intrinsic inhomogeneity [6,8–11]. Indeed, in a sequence of previous publications [12–14], it has been shown that the magnetic properties of manganites can be properly described within a mean-field approximation using Tsallis statistics.

2 Physical picture

We consider an inhomogeneous magnetic system composed by many homogeneous parts (clusters) with different sizes, each one of them described by the Maxwell-Boltzmann statistics. By averaging the magnetization over the whole system, we recover the Tsallis non-extensivity. A relationship between the q parameter and the moments of the distribution is obtained. The model is tested using Scanning Tunnelling Spectroscopy (STS) conductance maps, where the q parameter could be obtained and, consequently, the bulk magnetization predicted. We also apply the results to describe the magnetic behavior of $Cu_{90}Co_{10}$ amorphous ribbons.

The starting point is a magnetic system formed by small regions, or clusters of Maxwell-Boltzmann bits, each one of them with magnetization $\mathcal{M}(\mu, T, H)$ given by the usual Langevin function [15]:

$$\mathcal{M}(\mu, T, H) = \mu \left[\coth x - \frac{1}{x}\right] \tag{1}$$

where $x = \mu H/kT$. The clusters are distributed in size, and therefore in their net magnetic moment. Let $f(\mu)$ be the distribution of the clusters magnetic moment. Thus, the average magnetization of the sample will be given by:

$$\langle \mathcal{M} \rangle = \int_0^\infty \mathcal{M}(\mu, T, H) f(\mu) d\mu. \tag{2}$$

On the other hand, references [14,16] shows that the non-extensive magnetization is given by the generalized Langevin function:

$$\mathcal{M}_q = \frac{\mu_{ne}}{(2-q)} \left[\coth_q x - \frac{1}{x}\right] \tag{3}$$

where $q \in \Re$ is the Tsallis entropic parameter and $x = \mu_{ne} H/kT$.

Equating the susceptibilities

$$\chi_q = \langle \chi \rangle \equiv \lim_{H \to 0} \frac{\partial M}{\partial H} \tag{4}$$

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and the saturation values ($H \to \infty$) of equations (2) and (3), we find a analytical expression to the q parameter:

$$q(2-q)^2 = \frac{\langle \mu^2 \rangle}{\langle \mu \rangle^2} \quad (5)$$

where $\langle \mu \rangle$ and $\langle \mu^2 \rangle$ are the first and second moments of the distribution $f(\mu)$, respectively. This result is valid for any $f(\mu)$, and is analogous to that obtained by Beck [17] and Beck and Cohen [18].

Now, let us consider a distribution of critical temperatures. Again, the magnetization of a small region is given by the usual Langevin function (Eq. (1)), however, within the mean-field approximation, where:

$$x = \frac{\mu(H_0 + \lambda \mathcal{M})}{kT} \quad (6)$$

and $\lambda = 3kT_C/\mu^2$ corresponds to the mean-field parameter. Unfortunately, considering a distribution of critical temperatures, the average magnetization

$$\langle \mathcal{M} \rangle = \int_0^\infty \mathcal{M}(\mu, T, H_0, T_C, \mathcal{M}) f(T_C) dT_C \quad (7)$$

can not be reduced in a similar fashion to what was done in the case of a distribution of magnetic moments. Thus, a closed and simple expression connecting the q parameter and the moments of the distribution of critical temperatures could not be obtained. However, as will be discussed, the connection with experimental results will provide some suggestions.

3 Connections with experimental results

3.1 Distribution of magnetic moments

As discussed before, the q parameter is related to the moments of the distribution of magnetic moments (Eq. (5)). Thus, to measure the entropic parameter we need to measure, a priori, the distribution of magnetic moments. Below, we will discuss how to extract $f(\mu)$ from some experimental data.

3.1.1 Manganites

Colossal magnetoresistance (CMR) effect [8], usually observed on manganites, has been proposed in terms of intrinsic inhomogeneities [6,8,9], which can lead to a formation of insulating and conducting domains within a single sample, i.e., electronic phase separation in a chemical homogeneous sample. The inhomogeneities alter the local electronic and magnetic properties of the sample and should therefore be visible via STS [9,19,20] or Magnetic Force Microscopy (MFM) [21,22].

Becker and co-workers [9] measured STS in a $La_{0.7}Sr_{0.3}MnO_3$/MgO thin film and visualized a domain structure of conducting (ferromagnetic) and insulating

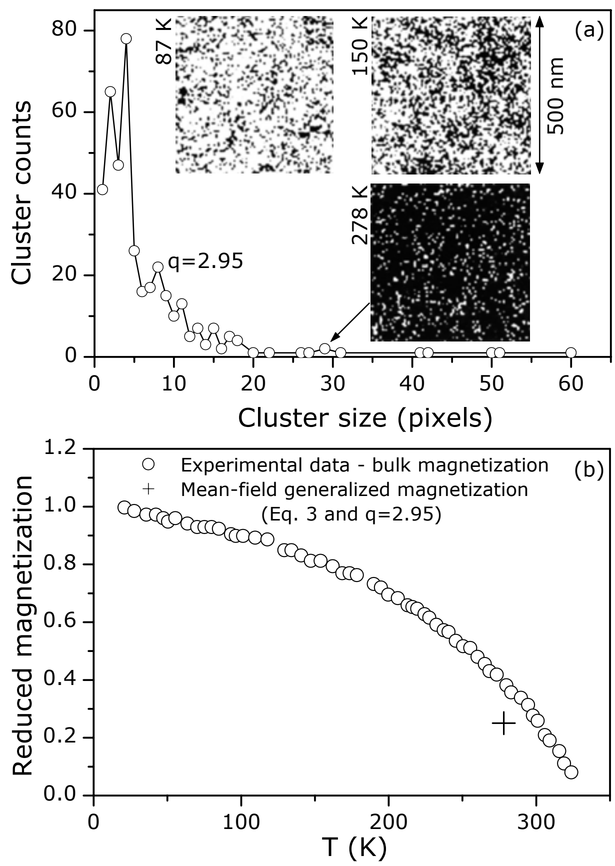

Fig. 1. (a) Scanning tunnelling spectroscopy images obtained at different temperatures on $La_{0.7}Sr_{0.3}MnO_3$/MgO manganite thin film, after Becker et al. [9]. White regions are conducting (ferromagnetic phase) clusters and the black regions are insulating clusters (paramagnetic phase). The main graphic is the cluster size distribution of the image at 278 K, proportional to the cluster magnetic moment distribution, where, using equation (8), $q = 2.95$ could be directly obtained. (b) Using the mean-field (reduced) generalized magnetization (Eq. (3): + symbol), we could predict the bulk magnetization, in a satisfactory agreement with the measured one (○ symbol), in a reduced scale $\mathcal{M}/\mathcal{M}$ (18 K) [9].

(paramagnetic) regions with nanometric size, since this manganite has a transition from a metallic phase (below T_C) to an insulating phase (above T_C), with a strong phase coexistence/competition around $T_C \sim 330$ K. These STS conductance maps obtained by those authors at 87 K, 150 K and 278 K are reproduced in Figure 1a. From these 1-bit images (black regions mean insulating/paramagentic phase and white regions stand for conducting/ferromagnetic phase), it was possible to determine the distribution of clusters size. Considering that the cluster size ϕ, measured in *pixels*, is proportional to the magnetic moment μ of the cluster, equation (5) can be re-written as:

$$\frac{\langle \phi^2 \rangle}{\langle \phi \rangle^2} = \frac{\langle \mu^2 \rangle}{\langle \mu \rangle^2} = q(2-q)^2. \quad (8)$$

The conductance map at 278 K has a distribution of clusters as presented in Figure 1a, and, using equation (8) we obtained from the data $q = 2.95$.

With this value of q, the total magnetization of the system can be predicted, by considering the mean-field approximation into the generalized magnetization (Eq. (3)), where $x = 3m_q/t$, $m_q = \mathcal{M}_q/\mu_{ne}$, $t = T/T_C^{(1)}$ and $T_C^{(1)} = 298$ K [12] (see Refs. [12,14] for details concerning the mean-field approximation applied to the non-extensive magnetization). This procedure results in a satisfactory agreement between the predicted reduced magnetization ($m_q = 0.25$; the + symbol) and the experimental one (the $\circ$ symbol), obtained measuring the bulk magnetization [9], as presented in Figure 1b. The images at 87 K and 150 K were not analyzed, since the clusters have already percolated.

The procedure above described shows how to extract the q parameter from an experimental data, and then how to apply the obtained q parameter to predict macroscopic quantities of the system. In addition, these results exemplify the relation between non-extensivity and microscopic inhomogeneities. Finally, it is important to stress that q is related to the dynamics of the system, since it measures the distribution of magnetic moments, that contains the dynamics.

3.1.2 Granular Alloys

Ferrari and co-workers [23] analyzed some melt-spun $Cu_{90}Co_{10}$ ribbons, materials intrinsically inhomogeneous [24,25]. They considered equation (2) to study the magnetic behavior of these materials, supposing a Log-Normal distribution of magnetic moments:

$$f(\mu) = \frac{1}{\sqrt{2\pi}\; s\; \mu}\; \exp\left[-\frac{\ln^2(\mu/\mu_0)}{2\; s^2}\right] \qquad (9)$$

where the kth moment is $\langle\mu^k\rangle = \mu_0^k \exp(k^2 s^2/2)$.

Those authors used this model to fit the magnetization curves at room temperature, as displayed in Figure 2-top. The fitting parameters for the sample (a) are $\mu_0 = 500\mu_B$ and $s = 1.16$; and for the sample (b), $\mu_0 = 3900\mu_B$ and $s = 0.93$. Both samples are $Cu_{90}Co_{10}$, however, prepared under different conditions [23]. With these values and using equations (9) and (5), we could obtain the q parameter for both samples: (a) $q = 3.11$ and (b) $q = 2.90$.

With those values of q, the non-extensive magnetic susceptibility was obtained:

$$\chi_q = \frac{q\mu_{ne}^2}{3kT} = \frac{q(2-q)^2\langle\mu\rangle^2}{3kT} \qquad (10)$$

and matches the experimental one, as presented in Figure 2-bottom.

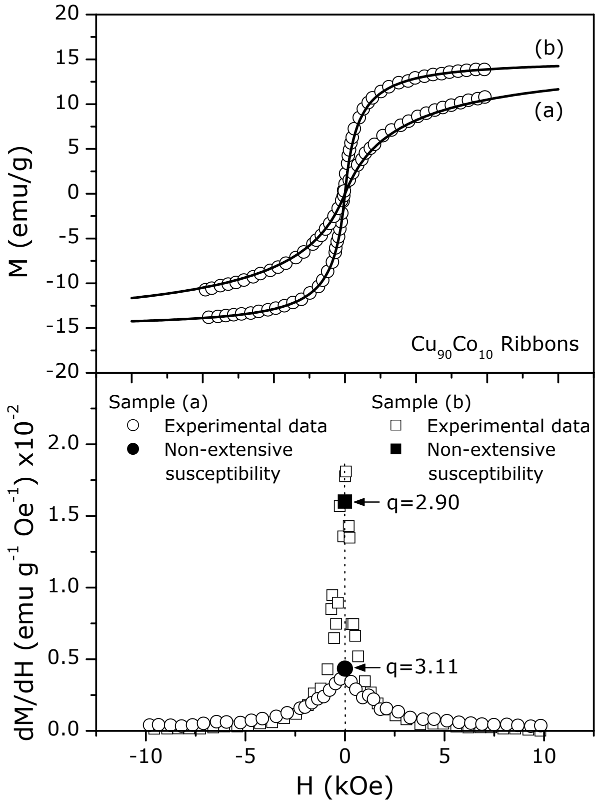

Fig. 2. Top: open circles are the experimental results, whereas the solid lines correspond to the average magnetization (Eq. (2)), considering a Log-Normal distribution (Eq. (9)). Bottom: dM/dH as a function of the applied magnetic field, where the magnetic susceptibility lies in the zero field limit. The predicted non-extensive magnetic susceptibility matches the experimental one, for both samples.

3.2 Distribution of critical temperatures

Campillo and co-workers [26] analyzed the magnetic properties of 200-nm thick films of $La_{0.67}Ca_{0.33}MnO_3$, which were grown under identical conditions onto five different single crystal substrates: MgO, Si, $NdGaO_3$, $SrTiO_3$, $LaAlO_3$. The films exhibit a strong substrate dependence of the magnetic properties, including, for instance, different values of Curie temperature T_C. On the other hand, the strain due to the growth mode induces inhomogeneities on the sample [11,26]. Campillo verified that these inhomogeneities depend on the substrate character and therefore they determined the T_C distribution for each thick film.

The authors considered that the magnetization of a small and homogeneous region is given by:

$$\mathcal{M}(m_0, T_C, T, \beta) = m_0 \left(1 - \frac{T}{T_C}\right)^{\beta} \theta(T_C - T) \qquad (11)$$

where $\theta(x)$ is the Heavyside Step function. Supposing a Normal distribution of Curie temperatures

$$f(T_C) = \frac{1}{\sqrt{2\pi}\; \sigma}\; \exp\left(-\frac{(T_C - \langle T_C\rangle)^2}{2\; \sigma^2}\right) \qquad (12)$$

where σ is the standard deviation and $\langle T_C\rangle$ the first moment of the distribution, they could write the average

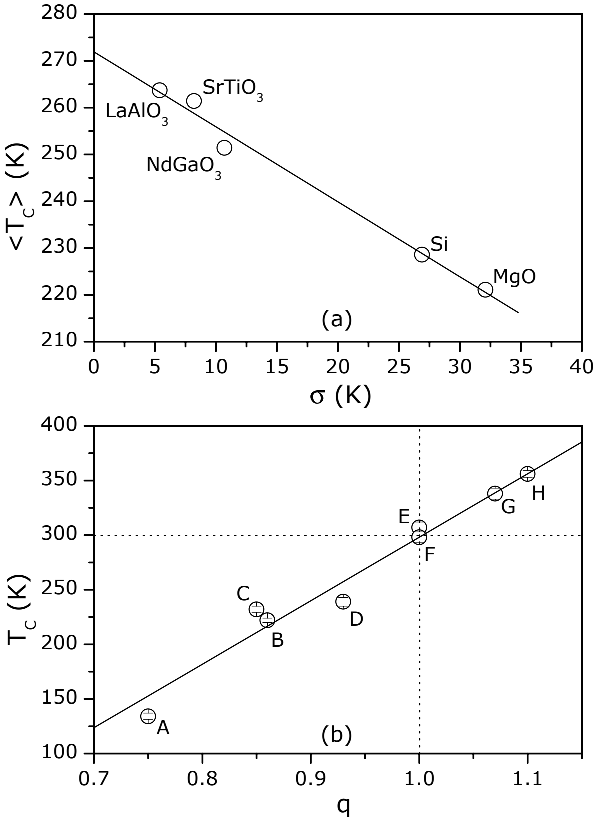

Fig. 3. (a) First moment $\langle T_C \rangle$ of the Curie temperature distribution as a function of the standard deviation σ, obtained fitting the M vs. T curves of $La_{0.67}Ca_{0.33}MnO_3$ manganites deposited onto five different single crystal substrates: MgO, Si, $NdGaO_3$, $SrTiO_3$, $LaAlO_3$. After Campillo et al. [26]. (b) Curie temperature as a function of the q parameter, obtained from the fit, using the generalized Brillouin function [12,13], of the M vs. T curves of those manganites referred in Table 1.

magnetization:

$$\langle \mathcal{M} \rangle = \int_0^\infty \mathcal{M}(m_0, T_C, T, \beta)\; f(T_C) dT_C \qquad (13)$$

and fit the corresponding M vs. T curves, for all samples available. From those fits, the authors could obtain a linear relationship between the standard deviation σ and the mean value $\langle T_C \rangle$ of the samples analyzed; $\langle T_C \rangle = 272 - 1.6\sigma$, as presented in Figure 3a.

On the other hand, in our previous work [12], we used the Generalized Brillouin function [12,13], a non-extensive magnetic equation of state, to fit the M vs. T curves obtained from those manganites presented in Table 1. From that study emerges a linear relationship between T_C, a quantity intrinsic to the material, and the q parameter, a quantity intrinsic to the non-extensive statistics; $T_C = -283 + 581\, q$, as presented in Figure 3b.

Comparing the results obtained by our own [12] (Fig. 3b) and Campillo [26] (Fig. 3a), we could estimate a relation between the standard deviation σ and the q parameter:

$$\sigma \propto (1 - q) \qquad (14)$$

Table 1. Manganites, and the respective references, in which its M vs. T curves were analyzed using the generalized Brillouin function [12,13] in order to obtain the corresponding q parameter. The Label column corresponds to the Figure 3b. The choice of compounds was made such as to cover a wide range of T_c values within the ferromagnetic phase, but it was random in any other aspect.

Label	Compound	Ref.
A	$La_{0.62}Y_{0.07}Ca_{0.31}MnO_{3+\delta}$	[27]
B	$La_{0.875}Sr_{0.125}MnO_{3+\delta}$	[28]
C	$La_{0.5}Ca_{0.5}MnO_3$	[29]
D	$La_{0.83}Sr_{0.17}Mn_{0.98}Fe_{0.02}O_3$	[30]
E	$La_{0.89}Sr_{0.11}MnO_{3+\delta}$	[12]
F	$La_{0.75}Ba_{0.25}MnO_3$	[31]
G	$La_{0.5}Ba_{0.5}MnO_3$	[31]
H	$La_{0.7}Sr_{0.3}Mn_{0.9}Ru_{0.1}O_3$	[32]

reinforcing the idea that the q parameter is related to the inhomogeneities of the system. Note that, $q \to 1$ imply in $\sigma \to 0$, i.e., the extensive (Maxwell-Boltzmann) limit corresponds to the homogeneous case.

4 Concluding remarks

Summarizing, in the present work we shown that the q parameter measures the inhomogeneity and dynamics of a given inhomogeneous magnetic system. We stress that the measured q, obtained from scanning tunnelling spectroscopy on manganites (a microscopic information), is able to predict a thermodynamic quantity, the bulk magnetization (a macroscopic information); the entropic parameter contains the dynamics, connecting the microscopic and macroscopic worlds. Thus, the present work shows a clear combination of dynamics and statistics, the key role to describe complex systems. The present model was also successful applied to a series of $La_{0.67}Ca_{0.33}MnO_3$ thick films [26] and melt-spun $Cu_{90}Co_{10}$ ribbons [23]; and the results reinforce the conclusions made in this work.

We acknowledge CAPES-Brasil, CNPq-Brasil and GRICES-Portugal. We are also thankful to P.B. Tavares and A.M.L. Lopes (sample preparation), and M.P. Albuquerque and A.R. Gesualdi (some discussion on image processing).

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