Experimental determination of the nonextensive entropic parameter q

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(Received 21 July 2005; published 2 March 2006)

We show how to extract the q parameter from experimental data, considering an inhomogeneous magnetic system composed by many Maxwell-Boltzmann homogeneous parts, which after integration over the whole system recover the Tsallis nonextensivity. Analyzing the cluster distribution of $La_{0.7}Sr_{0.3}MnO_3$ manganite, obtained through scanning tunneling spectroscopy, we measure the q parameter and predict the bulk magnetization with good accuracy. The connection between the Griffiths phase and nonextensivity is also considered. We conclude that the entropic parameter embodies information about the dynamics, the key role to describe complex systems.

DOI: 10.1103/PhysRevB.73.092401 PACS number(s): 05.70.Ce, 05.70.Fh, 75.47.Gk, 75.47.Lx

It is widely accepted that the statistical description of a system should be based on its dynamics; however, it is an information that indeed does not lie in the Boltzmann entropy. This fact opens a path for new and different statistics other than the Boltzmann one. In this direction, Tsallis thermostatistics has been strongly used in a number of different contexts.[1] This 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, seem 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, a sequence of previous publications[12–14] has shown that the magnetic properties of manganites can be properly described within a mean-field approximation using Tsallis statistics.

In the present Brief Report, through an analogy to the works of Beck[15] and Beck and Cohen,[16] we consider an inhomogeneous magnetic system composed by many homogeneous parts 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 nonextensivity. From this point of view, an analytical relationship between the q parameter and the moments of the distribution was obtained. The robustness of the present model was tested using scanning tunneling spectroscopy (STS) conductance maps, where the q parameter could be obtained and, consequently, the bulk magnetization predicted. In order to strengthen the connections between inhomogeneity and nonextensivity, it is shown that the description of manganites using Griffiths phase[17,18] is related to the nonextensive treatment, which contains the dynamics of the system.

We start considering a magnetic system formed by small regions, or clusters of Maxwell-Boltzmann bits, each of them with magnetization $\mathcal{M}$ given by the simple Langevin function.[19] The clusters are distributed in size, and therefore in their net magnetic moment. Thus let $f(\mu)$ be the distribution of magnetic moment of the clusters. The average magnetization of the sample will be given by

$$\langle \mathcal{M} \rangle = \int_0^\infty \mathcal{M} f(\mu) d\mu. \tag{1}$$

Our goal is to connect the above expression to the nonextensive magnetization that is calculated in Refs. 14 and 20:

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

where $x=\mu_{ne}H/kT$, $\coth_q$ is the generalized q-hyperbolic cotangent,[21] $q \in \mathrm{Re}$ is the Tsallis entropic parameter, and μ_{ne} means the magnetic moment of each nonextensive cluster. The nonextensive correlations lie inside each cluster, whereas the interactions interclusters remain extensive. This guarantees that the total magnetization will be additive.[14]

Microscopic analysis. From Eqs. (1) and (2), the average and nonextensive magnetic susceptibilities can be derived: $\langle\chi\rangle=\langle\mu^2\rangle/3kT$ and $\chi_q=q\mu_{ne}^2/3kT$, as well as the saturation values of the magnetization: $\langle\mathcal{M}\rangle_{sat}=\langle\mu\rangle$ and $\mathcal{M}_{q,sat}=\mu_{ne}/(2-q)$. Thus, equating those limits ($\langle\chi\rangle=\chi_q$ and $\langle\mathcal{M}\rangle_{sat}=\mathcal{M}_{q,sat}$), we find a microscopic analytical expression to the q parameter, in the sense that it is related to microscopic information (distribution of magnetic moments):

$$q(2-q)^2 = \frac{\langle\mu^2\rangle}{\langle\mu\rangle^2}, \tag{3}$$

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[15] and Beck and Cohen[16] in other contexts.

Macroscopic analysis. From Eq. (2) we can obtain a macroscopic analytical expression for the q parameter (similarly to what was done in Ref. 14), in the sense that it is now related to macroscopic quantities:

$$q(2-q)^2 = \frac{3kT\chi}{\mathcal{M}_{sat}^2}, \tag{4}$$

where χ and $\mathcal{M}_{sat}$ are the experimental magnetic susceptibility and saturation value, respectively. Note that for an ho-

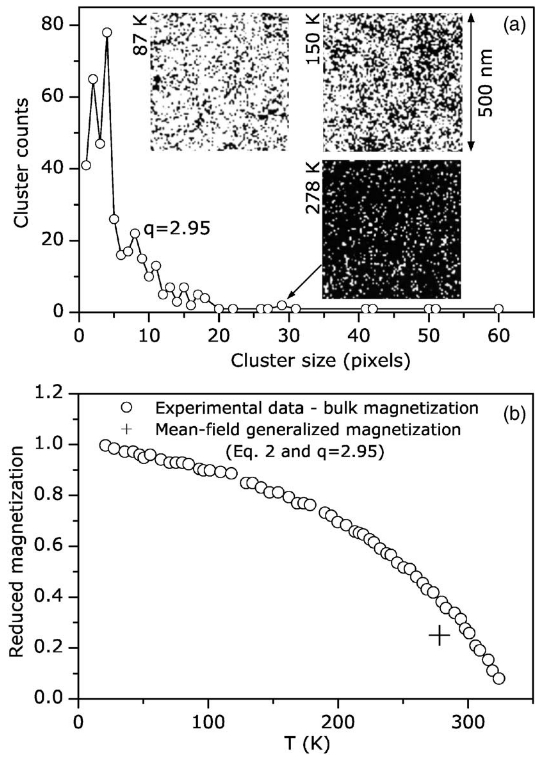

FIG. 1. (a) Scanning tunneling spectroscopy images on $La_{0.7}Sr_{0.3}MnO_3/MgO$ manganite thin film, after Becker *et al.* Ref. 9. White regions mean conducting (ferromagnetic) clusters and the black regions stand for insulating (paramagnetic) phase. The main graphic is the cluster size distribution of the image at 278 K, proportional to the cluster magnetic moment distribution, where, using Eq. (5), $q=2.95$ could be directly obtained. (b) Using the mean-field (reduced) generalized magnetization [Eq. (2): + symbol], we could predict the bulk magnetization, in a satisfactory agreement with the measured one (○ symbol), in a reduced scale $\mathcal{M}/\mathcal{M}(18\text{ K})$ Ref. 9.

mogenous superparamagnetic-like system, $q=1$.

Experimental connection with the microscopic analysis. The colossal magnetoresistance (CMR) effect,[8] usually observed on manganites, has been explained in terms of intrinsic inhomogeneities,[6,8,9] which lead to the formation of insulating and conducting domains within a single sample, i.e., electronic phase separation in a chemically homogeneous sample. The inhomogeneities alter the local electronic and magnetic properties of the sample and should therefore be visible via STS[9,22,23] or magnetic force microscopy (MFM).[24,25]

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 (paramagnetic) regions with nanometric size. 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. The STS conductance maps obtained by those authors at 87, 150, and 278 K are reproduced in Fig. 1(a). From these 1-bit images (black regions mean insulating/paramagentic phase and white regions stand for conducting/ferromagnetic phase), we have determined the distribution of clusters size. Considering that the cluster size ϕ, measured in *pixels*, is proportional to the magnetic moment μ of the cluster, Eq. (3) can be rewritten as

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

The conductance map at 278 K has a distribution of clusters as presented in Fig. 1(a), and, using Eq. (5), we obtain 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. (2)], where $x=3m_q/t$, $m_q=\mathcal{M}_q/\mu_{ne}$, $t=T/T_C^{(1)}$, and $T_C^{(1)}=298$ K (Ref. 12) (see Refs. 12 and 14 for details concerning the mean-field approximation applied to the nonextensive magnetization). This procedure results in a satisfactory agreement between the predicted reduced magnetization ($m_q=0.25$) and the experimental one, obtained measuring the bulk magnetization,[9] as presented in Fig. 1(b). The images at 87 and 150 K were not analyzed, since the clusters have already percolated.

The procedure described above shows how to extract the q parameter from 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 nonextensivity 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.

Experimental connections with the macroscopic analysis. $Pr_{0.05}Ca_{0.95}MnO_3$ ($T_C=110$ K), $LaMnO_3$ ($T_C=138$ K), and $La_{0.7}Ca_{0.3}MnO_3$ ($T_C=225$ K) are interesting examples of manganites. The inverse susceptibility as a function of temperature presents a strong downturn around T_C. These samples were prepared and measured as reported in Refs. 26 and 27, except the La-Ca manganite, where the experimental susceptibility was reproduced from Ref. 18.

The lower panels of Fig. 2 (□ symbols) present the q parameter obtained from the macroscopic analysis; using Eq. (4), the measured magnetic susceptibility (experimental data presented in the upper panels of Fig. 2) and the magnetic saturation value for the above cited manganites (Pr-manganite: 8.9 emu/g and La-manganite: 8.5 emu/g). This procedure was not done for the La-Ca manganite, since Ref. 18 does not provide the corresponding absolute values for the magnetic susceptibility.

Connections with Griffiths phase. An extensive number of papers Refs. 28–32, and references therein, deal with the dynamics of random magnetic systems, with special attention for a diluted Ising ferromagnet. The systems of interest are obtained by starting with an ordinary Ising model, which contains spins located on the vertices of a regular lattice. For a bond dilution, the nearest-neighbor interactions J_{ij} are independent random variables taking the values J and 0 with probabilities p and $1-p$, respectively. For a site dilution, $J_{ij}=Jc_ic_j$, where $c_{i,j}=1$ or 0, with probabilities p and $1-p$, respectively. This diluted ferromagnet is in the Griffiths phase[17,18,30–32] if its temperature is between the critical temperature $T_C(p)$ and the critical temperature $T_G=T_C(1)$ of a pure/nondiluted system. In Fig. 3, the dilution p-temperature T plane sketches this scenario.

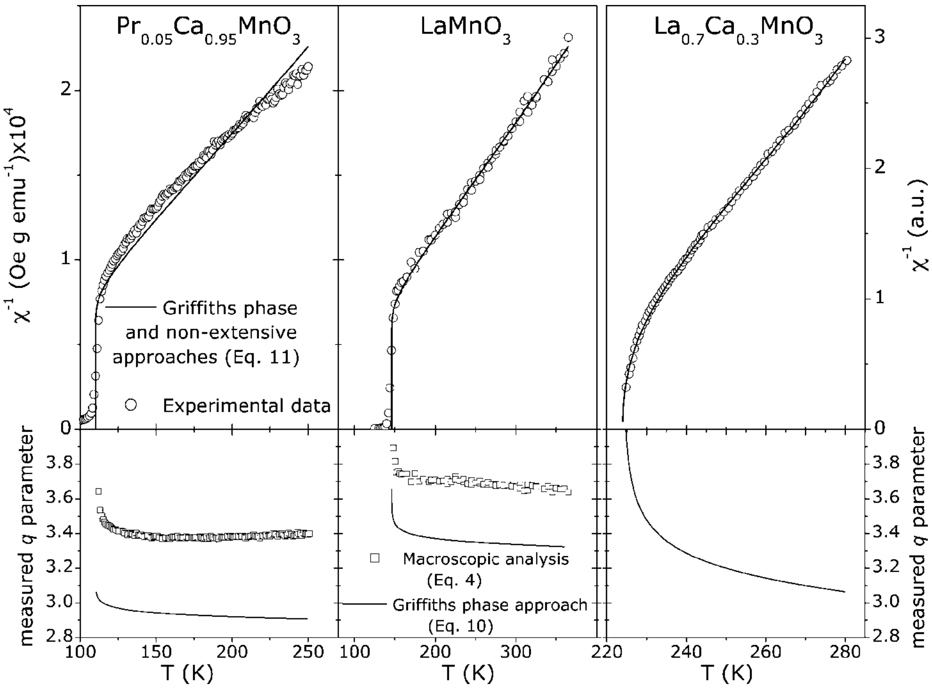

FIG. 2. Upper panels: Experimental (open circles) and theoretical [solid line—Eq. (11)] temperature dependence of the inverse susceptibility for the $La_{0.7}Ca_{0.3}MnO_3$ [after Salamon *et al.* Ref. 18], $LaMnO_3$, and $Pr_{0.05}Ca_{0.95}MnO_3$ manganites. Lower panels: measured q parameter from the macroscopic analysis and Griffiths phase approach.

Salamon and co-workers[17,18] have used the idea of Griffiths singularity to study manganites, considering a distribution of the inverse magnetic susceptibility λ:

$$f(\lambda) = \frac{A^{c-1}\lambda^{-c}\exp(-A/\lambda)}{\Gamma(c-1,A/T)} \tag{6}$$

to explain the sharp downturn in the $\langle\chi\rangle^{-1}(T)$ curve (behavior usually observed in manganites, as displayed in the upper panels of Fig. 2). In the expression above, c is a parameter of the distribution, Γ stands for the incomplete Gamma function, and

$$A = a\frac{\left(\frac{T}{T_C}-1\right)^{2(1-\beta)}}{\left(1-\frac{T}{T_G}\right)^{2\beta}}, \tag{7}$$

where a is a free parameter, $\beta=0.38$ is a critical exponent for the pure system, assumed to be three-dimensional Heisenberg-like, and T_G is the Griffiths temperature.[17,18,30–32] From Eq. (6) one can find the inverse average susceptibility:[33]

$$\langle\chi\rangle^{-1} = A\frac{\Gamma(c-1,A/T)}{\Gamma(c,A/T)} \tag{8}$$

that fits the strong downturn usually found on manganites.

The Curie law $\chi=\mu^2/3kT$ of a small Maxwell-Boltzmann region tells us that inhomogeneities in χ can arise from distributions of either μ or T (or both). Thus we can obtain from Eq. (6) a corresponding distribution of magnetic moments:

$$f(\mu) = \left(\frac{A}{3T}\right)^{c-1}\frac{2\mu^{2c-3}}{\Gamma(c-1,A/T)}\exp\left(-\frac{A\mu^2}{3T}\right) \tag{9}$$

and, consequently, the q parameter:

$$\frac{\langle\mu^2\rangle}{\langle\mu\rangle^2} = \frac{\Gamma(c,A/T)\Gamma(c-1,A/T)}{\Gamma(c-1/2,A/T)^2} = q(2-q)^2 \tag{10}$$

This result shows that, since $\langle\mu\rangle$ and $\langle\mu^2\rangle$ are both temperature dependent, one can expect that q will also be T-dependent. This is expected, since the distribution of magnetic moments changes as a function of temperature, changing also the q parameter. Indeed, it can be observed in the STS images presented in Fig. 1(a), reinforcing the idea that q is related to the dynamics of the system.

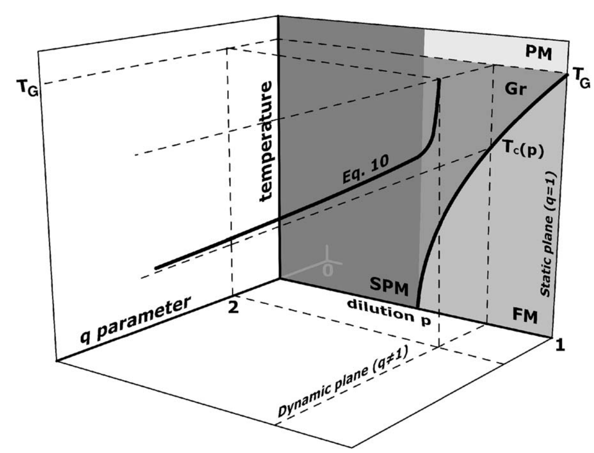

FIG. 3. Connections between Griffiths phase and nonextensivity, in a q-p-T space. p-T plane: usual magnetic phase diagram for a site/bond diluted magnetic system, where SPM is the superparamagnetic-like phase, PM the paramagnetic phase, FM the ferromagnetic phase, and Gr the Griffiths phase. This plane contains the critical temperature T_C and dilution p, basic (and static) information about the system. The q-T plane contains information related to the dynamics of the system, through Eq. (10). See text for details.

From the above, one can write the nonextensive magnetic susceptibility:

$$\chi_q = \frac{q\mu_{ne}^2}{3kT} = \frac{q(2-q)^2\langle\mu\rangle^2}{3kT} = \frac{\langle\mu^2\rangle}{3kT} = \langle\chi\rangle \tag{11}$$

that is equal to the average one. It is important to note that, once we know $f(\mu)$, the value of q can be directly obtained, and, consequently, χ_q. The present approach, as well as those presented previously, *does not* consider the entropic index q as a fitting parameter, but as a known quantity, previously determined and directly related to the inhomogeneity and dynamics of the system.

The model described above [Eq. (11)] is presented in the upper panels of Fig. 2. To obtain those results, the distribution of magnetic moments $f(\mu)$ presented in Eq. (9) has the following parameters: $c=-0.04$, $a=0.002$ K, and $T_G=510$ K, for $Pr_{0.05}Ca_{0.95}MnO_3$; $c=0.01$, $a=4.7\times10^{-8}$ K, and $T_G=555$ K, for $LaMnO_3$; and $c=0.32$, $a=0.066$ K, and $T_G=335$ K, for $La_{0.7}Ca_{0.3}MnO_3$. These parameters for the La-Ca manganite are considerable different from those presented in Ref. 18, due to the remarkable difference between the fit presented by those authors and that one sketched in Fig. 2. The entropic index q (lower panels of Fig. 2) *is not* a free parameter and could be obtained *a priori*, using Eqs. (3) and (9). It is important to stress the agreement between the *measured q parameter* using the macroscopic analysis and the present approach.

The connection between Griffiths phase and nonextensivity can be visualized considering the q parameter as another dimension; an independent variable in a q-p-T space, as sketched in Fig. 3. Analogously to the Curie temperature, a dilution p characterizes a diluted magnetic system, since these (static) quantities are unique for a certain system. If we know only p and $T_C(p)$, it is not possible to achieve the dynamics of this diluted magnetic system. For instance, given a dilution p, and consequently a Curie temperature $T_C(p)$, there are $\binom{N}{pN}$ different ways to dispose these spins, considering a site dilution problem with N available positions in the lattice. Different distributions of spins imply different dynamics and, consequently, different macroscopic quantities, like magnetization. In this sense, these quantities [p and $T_C(p)$] are static. On the other hand, the results presented in this Brief Report shown how the q parameter is related to the dynamics of the system and, once known q, the dynamics is consequently determined. Thus we propose that the p-T plane at $q=1$ is a static plane, whereas the q-T plane [Eq. (10)], for a certain value of p [or $T_C(p)$] takes account the dynamics of the system.

Summarizing, in the present Brief Report we have shown that the q parameter measures the inhomogeneity and dynamics of a given inhomogeneous magnetic system. From the measured q, obtained from scanning tunneling spectroscopy on manganites (a microscopic information), one can predict a thermodynamic quantity, the bulk magnetization (a macroscopic information); the entropic parameter contains the dynamics, connecting the microscopic and macroscopic information. The present model was also successfully applied to other manganites (bulk and thin films) and melt-spun granular alloys, reinforcing the conclusions made.

We acknowledge CAPES-Brasil, CNPq-Brasil, and GRICES-Portugal. We are also thankful to P. B. Tavares, A. M. L. Lopes, M. P. Albuquerque, and A. R. Gesualdi for valuable discussions.

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