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Non-equilibrium thermodynamics and anomalous diffusion

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Abstract. The convenience of a new thermodynamic frame for the description of anomalous diffusion is explored. Our research, which makes use of a recent new definition for entropy arising from multifractal analysis, shows that both dynamical and thermodynamical effects may contribute to non-classical diffusion.

1. Introduction

This paper has a double motivation inspired in two different aspects of recent thermodynamics: on the one hand, some extensions of non-equilibrium thermodynamics to generalized transport equations [1–5] and, on the other hand, some attempts to define a non-additive entropy for multifractal systems [6–12]. Our aim here is to study the thermodynamic aspects of anomalous diffusion from these two different, although complementary, points of view.

Classically, the relation between irreversible thermodynamics and transport laws (Fick's, Fourier's laws, for instance) is well established [13, 14]. These laws have a wide range of validity and they are very useful for applications. However, there are situations in which they must be generalized taking into account, for instance, memory effects, non-local effects or nonlinear effects. The generalization of such transport laws has stimulated in recent years a corresponding generalization of the underlying non-equilibrium thermodynamics [1–5]. The basic idea of these developments is that dynamics and thermodynamics must be dealt with in parallel, rather than assuming that there is an *a priori* thermodynamics (namely, the local-equilibrium thermodynamics), which restricts the corresponding admissible dynamics. Thus, whereas some dynamic behaviours are incompatible with the local equilibrium hypothesis, it is possible to introduce non-classical entropies which are compatible with such dynamic behaviours [1, 2].

In parallel for the last 20 years, another trend in modern thermodynamics has focused on possible generalizations of the definition of entropy, which conserve their interpretation as lack of information but do not retain certain properties hitherto considered essential, such as extensiveness [6, 7, 15]. These new definitions apparently may apply to very complex systems with long-range interactions, persistent memory or systems evolving in a fractal space, where the intuitive reasons which justified extensiveness do not apply any longer. Furthermore, in these complex systems (for instance, gravitational systems, magnetic systems, random walks, etc), the usual thermodynamic formalisms fail whenever the relevant

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thermostatistical quantities are computed, which turn out to diverge. Here we will consider the definition proposed by Tsallis in 1988 [6] which has been seen to be a consistent generalization both of thermodynamics and of statistical mechanics [8]

$$S_q = -k \frac{1 - \sum_i p_i^q}{1 - q} \tag{1}$$

where k is the Boltzmann's constant, p_i the probability of microstate i and q a constant parameter. This generalized entropy reduces to the classical Boltzmann entropy in the limit $q \to 1$. Instances where this entropy has been successfully applied include stellar polytropes [10], with the parameter q being a function of the polytrope index, and Lévy walks [11], where q turns out to be related to the dimension of the resulting fractal trajectory.

In this paper, we deal with the thermodynamic aspects of non-Fickian diffusion. In contrast to classical diffusion, where the characteristic value r of the displacement (defined, for instance, as the root mean square of the displacement or as the radius of the sphere which contains 90% of the total number of particles) is proportional to $t^{1/2}$ in the long-time limit, in non-Fickian diffusion, or anomalous diffusion, it behaves as

$$r \sim t^{\sigma/2} \tag{2}$$

with $\sigma \neq 1$. If $\sigma = 1$ one recovers the usual diffusion, and for $\sigma < 1$, or $\sigma > 1$ one has subdiffusive or superdiffusive behaviour, respectively. Such non-Fickian aspects of diffusion have received much attention in the last decade [16–20], both because of their theoretical interest as well as for their applications (diffusion in fractal spaces, transport in ion conducting materials [17], flow in rocks [18], particle diffusion in fluctuating magnetic fields [19], transport in chaotic dynamics [20], etc). However, whereas the dynamic aspects of diffusion leading to (2) have deserved much interest, the thermodynamic aspects have not been studied in much detail. Only recently, some authors [11] have focused on the thermodynamic description of Lévy-like anomalous diffusion, even though most of the vast domain of anomalous diffusive phenomena remains virtually unexplored from the thermodynamic point of view.

Here we shall try to balance this situation by studying the convenience of the fractal entropy for the thermodynamic description of a kind of correlated anomalous diffusion, namely that arising from a nonlinear diffusion equation:

$$\frac{\partial u}{\partial t} = \nabla^2 u^\gamma. \tag{3}$$

This kind of equation has been seen to arise naturally when one considers the flow of homogeneous fluids through porous media [21], particle diffusion across magnetic fields [22] or the spatial spread of biological populations [23]. Very recently, this dynamics has been studied within the scheme of Tsallis' generalized statistical mechanics by resorting to the maximum entropy principle [12]. Here, the approach will be rather different, since our main aim is to establish whether a new thermodynamic frame is actually needed for this dynamics as far as non-equilibrium thermodynamics is concerned.

The paper is structured as follows: the second section contains a short overview of the standard thermodynamic derivation of the diffusion equation which then serves as a model to obtain the diffusion equation associated with the fractal entropy. In parallel, the diffusion transport coefficient is allowed to depend on the particle density, thus introducing some non-thermodynamic contributions to the anomaly in diffusion. The resulting equation is then solved and the consequences of the second principle on this dynamics are eventually discussed. In section 3 we study three particular cases and we finally expose the conclusions in section 4.

2. Fractal entropy and generalized diffusion equation

The classical approach to diffusion from thermodynamics, as may be found in classical references such as [13] or [24], starts from the usual definition for entropy as applied to a continuous distribution $P(x,t)$:

$$S(t) = -k \int P(\boldsymbol{x},t) \ln P(\boldsymbol{x},t)\, \mathrm{d}^N \boldsymbol{x} \tag{4}$$

By taking the continuity equation $\partial P/\partial t = -\nabla \cdot \boldsymbol{J}$ and assuming a linear relation between the flux $\boldsymbol{J}$ and the thermodynamic force $\nabla(\delta S/\delta P)$ in the form

$$\boldsymbol{J} = L\nabla \frac{\delta S}{\delta P} \tag{5}$$

the following diffusion equation emerges

$$\frac{\partial P}{\partial t} = k\nabla\left(L\nabla \ln P\right). \tag{6}$$

Then, it is generally assumed that the transport coefficient L in (5) is a constant times the distribution function P:

$$L = \frac{D}{k}P.$$

This is a subtlety in the derivation that we are especially interested in noting since it will be important later in this paper.

By using this relation one is finally able to cast equation (6) into the more familiar diffusion equation

$$\frac{\partial P}{\partial t} = D\nabla^2 P. \tag{7}$$

We now turn to a non-standard thermodynamics and we will study whether a non-classical definition of entropy can be related to non-classical diffusion. Instead of the usual Boltzmann definition for the entropy, namely

$$S_1 = -k\sum_i p_i \ln p_i \tag{8}$$

where k is Boltzmann's constant and p_i the probability of microstate i, we consider expression (1) proposed by Tsallis [6]. The motivation of (1) is to be found in measures arising in analysis of multifractals, and it has been used in several contexts by a number of authors in the last few years [8, 9, 11, 12]. Making use of the fact that $\sum_i p_i = 1$, one may rewrite definition (1) as

$$S_q = -\frac{k}{1-q}\sum_i p_i(1-p_i^{q-1}) \tag{9}$$

which clearly shows its positivity. A thorough analysis of the mathematical properties of (9) may be found in [7].

Here, we generalize (9) to a continuous distribution function $P(\boldsymbol{x},t)$ as

$$S_q(t) = -\frac{k}{1-q}\int P(\boldsymbol{x},t)[1-P^{q-1}(\boldsymbol{x},t)]\, \mathrm{d}^N \boldsymbol{x} \tag{10}$$

where $\boldsymbol{x}$ is the position in an N-dimensional space and t is the time.

By means of the formal structure of classical irreversible thermodynamics [13], as we have seen at the beginning of this section, one may identify the flux of probability in this generalized thermodynamic formalism as

$$\boldsymbol{J} = L\nabla\frac{\delta S_q}{\delta P} = \frac{kq}{1-q}L\nabla P^{q-1} \tag{11}$$

where L is a transport coefficient. When (11) is combined with the conservation of probability equation

$$\frac{\partial P}{\partial t} = -\nabla\cdot\boldsymbol{J} \tag{12}$$

one is led to the following diffusion equation for processes evolving under this thermodynamic frame:

$$\frac{\partial P}{\partial t} = -\frac{kq}{1-q}\nabla(L\nabla P^{q-1}). \tag{13}$$

It is worth noting that this kind of dynamics has a positive entropy production, as may be seen by evaluating the time derivative of S_q, namely

$$\frac{\mathrm{d}S_q}{\mathrm{d}t} = \frac{kq}{1-q}\int P^{q-1}\frac{\partial P}{\partial t}\,\mathrm{d}^N\boldsymbol{x}. \tag{14}$$

By introducing (13), integrating once by parts and assuming that the surface term vanishes one obtains

$$\frac{\mathrm{d}S_q}{\mathrm{d}t} = \left(\frac{kq}{q-1}\right)^2\int L(\nabla P^{q-1})^2\,\mathrm{d}^N\boldsymbol{x} \tag{15}$$

which, provided $L > 0$, is always positive or zero. This is only so, however, as long as the surface term in the derivation of (15) vanishes, namely when

$$LP^{q-1}\nabla P^{q-1}\big|_{\Sigma} = 0 \tag{16}$$

where Σ stands for the boundary of the volume of space available for diffusion (from now on we will take this volume to be unbounded). This relation will prove useful later in this paper to restrict the range of the possible values for the parameters of the scheme.

We now note that, since in classical irreversible thermodynamics one has $L = DP/k$ with D a constant, we are here generally led to accept a dependence for L such as $L = DP^{\alpha}/k$ with D and α constants, and k the Boltzmann constant. In the case $\alpha = 1$ one recovers the classical picture and for $\alpha \neq 1$ anomalous diffusive effects of a dynamic nature are introduced into the scheme. A concentration dependent diffusivity of this type has been proposed for fractal diffusion (see [25], and references therein), for particle diffusion across magnetic fields [22] and for the motion of a polytropic gas in a porous medium [21]. We will henceforth study the effects of the parameters q and α in the description of anomalous diffusion and thus discuss the convenience of a thermodynamic ($q \neq 1$) or a dynamic ($\alpha \neq 1$) description of the phenomenon, respectively.

Introducing the relation $L = DP^{\alpha}/k$ into equation (13) and using the fact that $P^{\alpha}\nabla P^{q-1} = (q-1)P^{\alpha+q-2}\nabla P = (q-1)(\alpha+q-1)^{-1}\nabla P^{\alpha+q-1}$, we obtain the following generalized diffusion equation,

$$\frac{\partial P}{\partial t} = D\frac{q}{\alpha+q-1}\nabla^2 P^{\alpha+q-1} \tag{17}$$

which is a nonlinear equation of the form advanced in (3), elsewhere referred to as the ‘porous media equation’. This equation has been thoroughly studied in several mathematical works [22, 26, 27]. Here we just quote the Barenblatt–Pattle solution. This is the physical solution for an initial delta distribution $P(\boldsymbol{x}, t) = \delta^N(\boldsymbol{x})$ and has the remarkable property of being the asymptotic $t \to \infty$ limit distribution of the solutions of (17) with a localized initial distribution $P(\boldsymbol{x}, 0)$ [27]. Therefore, this solution plays the same role in the dynamics (17) as the Gaussian does in the standard linear diffusion. Assuming $q > 0$, the solution of (17) is

$$P(\boldsymbol{x}, t) = bt^{-\mu} \left\{ \left[a^2 - x^2 t^{-2\mu/N} \right]_+ \right\}^{1/(q+\alpha-2)} \qquad \text{for } q + \alpha > 2 \tag{18}$$

$$P(\boldsymbol{x}, t) = bt^{-\mu} \left(\frac{1}{a^2 + x^2 t^{-2\mu/N}} \right)^{1/(2-q-\alpha)} \qquad \text{for } 2\frac{N-1}{N} < q + \alpha < 2 \tag{19}$$

where

$$b = \left(\frac{|q + \alpha - 2|}{2Dq[N(q + \alpha - 2) + 2]} \right)^{1/(q+\alpha-2)} \qquad \mu = \frac{N}{N(q + \alpha - 2) + 2}$$

$[\cdot]_+ = \max(\cdot, 0)$ and a is a constant (depending on q and α) to be determined by normalization. We may now advance the proposition that these solutions correspond to the subdiffusion and superdiffusion cases, respectively, as we shall substantiate in the following paragraph. The solutions with $q + \alpha < 2(N - 1)/N$ or $q < 0$ correspond to non-physical probability distributions (non-normalizable) and we, therefore, ignore them altogether in our further developments. A similar restriction, namely $q > 0$, was imposed in [7] on the grounds of purely mathematical arguments.

An important feature of these solutions is that they are of the form

$$P(x, t) = t^{-\mu} f(x t^{-\mu/N}) \tag{20}$$

and, therefore, their characteristic scaling may be easily seen to correspond to anomalous diffusive behaviour: if one takes a measure $r(t)$ of the spread of $P(\boldsymbol{x}, t)$ to be the radius of the sphere which contains a fraction $\beta < 1$ of the total probability

$$\int_0^{r(t_0)} P(|\boldsymbol{x}|, t_0) \Omega_N |\boldsymbol{x}|^{N-1} \, \mathrm{d}|\boldsymbol{x}| = \beta \tag{21}$$

where Ω_N is the surface of the N-dimensional sphere of unitary radius and then consider (21) again for a different time t, the change of variable $x' = (t/t_0)^{-\mu/N} |\boldsymbol{x}|$ leads one to

$$\int_0^{(t/t_0)^{-\mu/N} r(t)} P(x', t_0) \Omega_N x'^{N-1} \, \mathrm{d}x' = \beta. \tag{22}$$

A simple comparison of (21) and (22) leads to the conclusion that

$$r(t) = \left(\frac{t}{t_0} \right)^{\mu/N} r(t_0) \tag{23}$$

independently of β, whence one recognizes the typical anomalous diffusive scaling

$$r \sim t^{\mu/N} = t^{1/(N(q+\alpha-2)+2)}. \tag{24}$$

From this last relation it is straightforward to identify the coefficient of anomalous diffusion as $\sigma = 2/(N(q+\alpha-2)+2)$ which is greater than unity (superdiffusion) as long as $q+\alpha < 2$ and, conversely, $\sigma < 1$ if $q + \alpha > 2$.

We must now study under what conditions solutions (18) and (19) are fully acceptable in the scheme that we have here presented, in the sense that they lead to a finite S_q and that they satisfy the assumption on which the positivity of the entropy production relies, namely condition (16). On the one hand, solution (18) is bounded and vanishes identically at infinity so that both the convergence of S_q and condition (16) are trivially satisfied. On the other hand, the asymptotic behaviour of (19) is $P \sim |\boldsymbol{x}|^{2/(q+\alpha-2)}$ whereas for S_q to be finite one must require that $P(\boldsymbol{x}, t)$ fades away faster than $|\boldsymbol{x}|^{-N/q}$ for $|\boldsymbol{x}| \to \infty$ and the fulfilment of condition (16) demands that $P \sim x^{(1-\gamma)/(2q+\alpha-2)}$ with $\gamma > 0$ in this same limit. Consequently, after elementary calculations we find that the finiteness of the entropy and its positivity in the superdiffusive case ($q + \alpha < 2$) impose the following condition relating the values of q and α:

$$\max\left(2\frac{N-1}{N}, 2\frac{N-q}{N}, 2(1-q)\right) < q + \alpha < 2 \qquad \text{and} \qquad q > 0. \tag{25}$$

For the sake of clarity, we represent in figure 1 the domain of the plane (q, α) where the physical values of the parameters of our scheme lie according to condition (25).

3. Some particular cases

Let us now consider two limiting cases. First, let us suppose that all the anomaly in diffusion is to be accounted for by dynamical mechanisms, so we then have that in our scheme $q = 1$ (standard thermodynamics) and $\alpha \neq 1$. It is easy to show that in this situation, by choosing α according to $\alpha = 1 + (2/N)\ (1/\sigma) - 1)$, one is able to reproduce any arbitrary anomalous diffusive behaviour:

$$r \sim t^{\sigma/2} \qquad 0 < \sigma < \infty.$$

This is easily seen by imagining the function $\sigma = 2/(N(q + \alpha - 2) + 2)$ represented on the z-axis of the domain in figure 1 and slicing the resulting surface by the plane $q = 1$.

The other limiting case which is interesting to explore is the situation in which anomalous diffusion is to be considered exclusively as a thermodynamic phenomenon. In this case $\alpha = 1$ (standard dynamics) and $q \neq 1$ with $q = 1 + (2/N)(1 - \sigma/\sigma)$. When we do this, we are led to a restriction on the range over which the coefficient of anomalous diffusion is allowed to vary. This can be seen graphically by slicing now the representation of $\sigma = 2/(N(q + \alpha - 2) + 2)$ over the domain in figure 1 along the plane $\alpha = 1$.

As a result, we see that if anomalous diffusion is to be accounted for purely on account of a fractal thermodynamics, we find that $\sigma < (N + 2)/2$. Therefore, a description of turbulence when $N \leqslant 3$ (as long as it is described with the kind of correlated dynamics considered here), where one typically has $r \sim t^{3/2}$ [28], should demand at least a dynamic contribution ($\alpha \neq 1$) to the non-classical thermodynamics.

Essentially, no other substantial differences arise in our scheme regarding the distribution functions or the dynamical equations when one considers a pure dynamic or thermodynamic origin of the anomaly in diffusion, since the parameters α and q enter almost everywhere in our scheme symmetrically as $q + \alpha$.

Consequently, it seems more natural to allow a coupling of both dynamic and thermodynamic aspects of diffusion, so that α and q are not to be considered independent of each other. They could, for instance, be related linearly as

$$\alpha = Cq + (1 - C)$$

which ensures that when $q = 1$ the standard picture of diffusion ($\alpha = q = 1$) is encountered. Furthermore, if we assume that $C > -1$, then it is easily proved that $q < 1$ corresponds to superdiffusion and $q > 1$ to subdiffusion. If, in contrast, we have $C < -1$, we get $q < 1$ ($q > 1$) related to subdiffusion (superdiffusion).

As we have done in the previous cases, we can now slice the surface $\sigma = 2/(N(q + \alpha - 2) + 2)$ defined over the domain of physical parameters (q, α) (see figure 1) along

FIG. 1

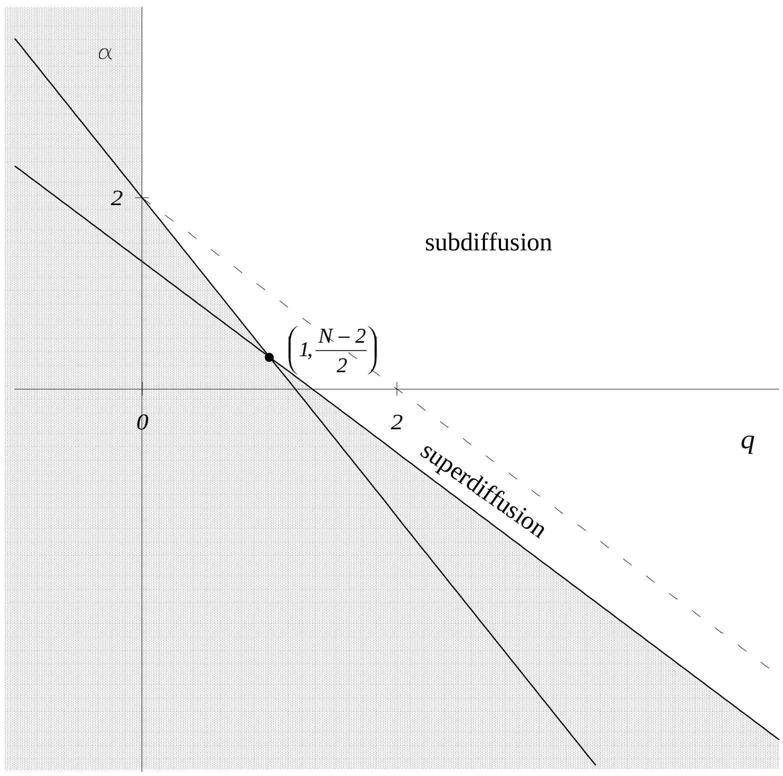

Figure 1. Distribution in the (q, α) plane of the domain of non-physical situations (shaded area), superdiffusion, subdiffusion and standard diffusion (dashed line) for a space of N dimensions.

the plane $\alpha = Cq + 1 - C$ for each of the above mentioned cases, namely $C > -1$ and

$C < -1$. One then finds that for $C > -1$ one can have $0 < \sigma < 1 + (N(C+1)/2)$ which means a restriction on superdiffusion, whereas for $C < -1$ one can reproduce dynamics subject to $2/(2 - N(C+1)) < \sigma < \infty$ and, therefore, only a limitation on subdiffusion is observed. It is, therefore, easy to see that the proposed combined effect of dynamics and thermodynamics can reproduce the typical behaviour of diffusion in fully developed turbulence, where $r \sim t^{3/2}$, as long as $C < -1$ or $C > (4/N) - 1$, and even account for intermittency effects [29].

4. Concluding remarks

Here we have studied the relation of a non-classical entropy with anomalous diffusion. The need for a generalization of the entropy to describe some kinds of non-Fickian effects was also pointed out in [30], in the very different context of case 2 diffusion, where relaxational effects must be taken into account in the diffusion flux. The relation pointed out here goes farther than in [30], where the relaxational effects implied a perturbative correction to the dynamics and the thermodynamics, which was of interest for short times. In the present work, the modification is deeper, both in the dynamics, because they are important not only in the short-time regime but also for long times, and because, as we have shown here, it might lead to a redefinition of the entropy rather than to a perturbative correction to it.

The relation we have shown points to a new possible application of the fractal entropy and reveals the intimate relation between dynamics and thermodynamics in a phenomenon of wide physical interest. In this paper we have seen that dynamic and thermodynamic aspects of diffusion are dynamically indistinguishable except for limitations on the speed of superdiffusion when an anomalous thermodynamics is present. An important result of this paper has been to show that under this scheme fractal thermodynamics cannot account alone for the superdiffusive properties of turbulence. Turbulence must necessarily include dynamic arguments, combined or not with an underlying non-standard thermodynamic frame.

To proceed further with the role of thermodynamic effects in anomalous diffusion, it could be enlightning to explore the equilibrium properties of a system under such a thermodynamic description. A correlation between equilibrium properties and anomalous diffusion set through the generalized entropy could help us elucidate the extent of the thermodynamic contribution to anomalous diffusion. It could be interesting as well to explore if anomalous diffusion in a shear flow leads to differences when one assumes a dynamic or a thermodynamic basis of diffusion. If it were so, this would provide us with an experimental procedure to establish which scheme suits better the description of a particular anomalous diffusive phenomenon and what values of q or α appropriately describe it.

On the other hand, our work points out the convenience of paying more attention to the thermodynamic aspects of non-Fickian diffusion which have been up to now practically ignored. A more detailed knowledge of the entropy could be useful when studying problems related to the dissipation in fractal sets, or the couplings of heat and mass transport, instead of focusing exclusively on mass transport.

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