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Answer to an open problem proposed by E Barkai and J Klafter

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Abstract

In a negative answer to the open problem proposed by E Barkai and J Klafter, it is proved that the theory presented by Amblard *et al* is not consistent with the GER. We suggest applying a theory in terms of a fractional Fokker–Planck equation to model the experiment measured by Amblard *et al*. The result obtained is consistent with the statement by Amblard *et al* (1996 *Phys. Rev. Lett.* **77** 4470) that the numerical pre-factors of the power law would be modified by local geometry and are not exact.

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1. Introduction

In a recent experiment Amblard *et al* [1] measured anomalous transport properties of magnetic beads embedded in a three-dimensional polymer network. In their paper, Amblard *et al* suggested a theory describing the motion of the beads.

Amblard *et al* found that when the beads are subjected to an external uniform force $\vec{F}$, the drift follows

$$\langle x_{\parallel}(t)\rangle_F \sim t^p, \qquad \text{with} \quad p = 0.76 \pm 0.03, \tag{1}$$

where $x_{\parallel}$ is the component of $\vec{x}$ along $\vec{F}$. In addition, they showed that the response to the external bias is linear. The authors also measured the diffusion of the beads in the absence of the field, and found

$$\left\langle x_{\parallel}^2(t)\right\rangle_0 \sim t^q, \qquad \text{with} \quad q = 0.73 \pm 0.01. \tag{2}$$

These observed anomalous power laws (1) and (2) can be readily explained if we consider the bead deforming the filaments of the cage surrounding it. Indeed, let us consider such a filament with bending constant κ, in a solvent of viscosity η. If s is the internal curvilinear coordinate along the polymer and $r(t,s)$ the transverse deformation of the filament, then the equation of movement for $r(t,s)$ is

$$\eta\frac{\partial r}{\partial t}=\kappa\frac{\partial^4 r}{\partial s^4}+f(t,s), \tag{3}$$

where $f(t,s)$ is the force acting on the filament. It is straightforward to show that the Green function, $F(t,s)$, associated with this equation can be written in the following scaling form:

$$F(t,s)=\frac{1}{\eta s}F\left(\frac{\eta s^4}{\kappa t}\right)\equiv\frac{1}{\eta^{3/4}\kappa^{1/4}t^{1/4}}\tilde{F}\left(\frac{\eta s^4}{\kappa t}\right). \tag{4}$$

If, for the sake of simplicity, we assume that the magnetic bead applies a constant point force, f at $s=0$, then the displacement of the centre of mass of the bead is given by

$$\langle x\rangle_f\approx\langle r(t,0)\rangle_f=f\int_0^t F(t-t',0)\,\mathrm{d}t'\approx\frac{4}{3}f\frac{\tilde{F}(0)t^{3/4}}{\eta^{3/4}\kappa^{1/4}}. \tag{5}$$

In the absence of the external force, the bead movement is dominated by the thermal motions of the surrounding filaments to which it is coupled. In this case, the mean-square displacement of the bead is the mean-square displacement of the filament,

$$\langle x^2\rangle_0=\langle r^2(t,s)\rangle_0=k_BT\eta\int\mathrm{d}s'\int_0^t F^2(t-t',s-s')\,\mathrm{d}t'\propto\frac{k_BTt^{3/4}}{\eta^{3/4}\kappa^{1/4}}. \tag{6}$$

It is interesting to note that, when one evaluates the numerical pre-factors in (5) and (6), one obtains a semi-quantitative agreement with known values of the f-actin rigidity [2]. The power laws of the time dependence described by (5) and (6), $t^{3/4}$, are not altered if one considers the bead interacting simultaneously with several filaments of the cage. The details of local geometry should modify numerical pre-factors in these theoretical formulae; undoubtedly, the bead has to push against several filaments simultaneously while the confined geometry must change the effective friction constants involved. In [3], Barkai and Klafter pointed out that although the validity of the generalized Einstein relation (GER) has not been discussed by the authors of [1], their measurements provide a direct opportunity to check this relation. When analysing the corrected results of [1] one finds that to a good approximation (equation (7)), the GER [4]

$$\left\langle x_{\parallel}^2(t)\right\rangle_0=\frac{2k_BT\langle x_{\parallel}(t)\rangle_F}{|\vec{F}|} \tag{7}$$

is valid. They conclude that the GER is well suited to describe the anomalous transport properties. They could not conclude whether the theory presented by Amblard *et al* is consistent with the GER. It would be interesting to clarify this issue. In section 2, we answer this open problem. In section 3, we give a theory to the GER.

2. Answer to the above open problem

In this section, we discuss the open problem, i.e., whether the theory presented by Amblard *et al* [1] is consistent with the GER. From the second equality of equation (4), we have

$$F(\zeta)=\zeta^{\frac{1}{4}}\tilde{F}(\zeta), \tag{8}$$

where $\zeta=\eta s^4/\kappa t$. Since $\tilde{F}(0)=\lim_{\zeta\to 0}[F(\zeta)/\zeta^{1/4}]$ and $\tilde{F}(0)\neq 0$, for any given $\varepsilon>0$ there exists a positive number $\delta=\delta(\varepsilon)$ related with ε such that for all ζ, $|\zeta|<\delta$, we have

$$F(\zeta)\approx\tilde{F}(0)\zeta^{1/4}. \tag{9}$$

Hence,

$$F(t,s) \approx \frac{\tilde{F}(0)}{\eta^{3/4}\kappa^{1/4}} t^{-\frac{1}{4}} \tag{10}$$

for all s such that $|s| < \left(\frac{\kappa\delta}{\eta}\right)^{1/4} t^{1/4}$. From (6) and (10), a simple calculation yields

$$\langle x^2 \rangle_0 \approx 2k_B T \frac{s\tilde{F}^2(0)}{\eta^{1/2}\kappa^{1/2}} t^{1/2} \tag{11}$$

for $|s| < \left(\frac{\kappa\delta}{\eta}\right)^{1/4} t^{1/4}$. This implies that

$$\langle x^2 \rangle_0 < 2k_B T \delta^{\frac{1}{4}} \frac{\tilde{F}^2(0)}{\eta^{3/4}\kappa^{1/4}} t^{3/4}. \tag{12}$$

From (5) and (12), finally we have

$$\langle x^2 \rangle_0 < \frac{3\tilde{F}(0)}{4} \delta^{\frac{1}{4}} \left[\frac{2k_B T}{f} \langle x \rangle_f \right] \tag{13}$$

for any give ε. Since $\delta = \delta(\varepsilon) \to 0$ when $\varepsilon \to 0$, then for any give $\rho < 1$ and for sufficiently small ε, we have

$$\langle x^2 \rangle_0 < \rho \frac{2k_B T}{f} \langle x \rangle_f. \tag{14}$$

But we know that $\langle x^2 \rangle_0 = \frac{2k_B T}{f} \langle x \rangle_f$ [3]. This shows that the theory of [1] is not consistent with the GER.

3. GER for FFPE

In [5], in order to describe anomalous systems close to thermal equilibrium based on fractional derivatives, Metzler, Barkai and Klafter presented a one-dimensional fractional Fokker–Planck equation (FFPE):

$${}_0D_t^{\alpha} W(x,t) = G L_{\mathrm{FP}} W(x,t), \tag{15}$$

where $W(x,t)$ is the probability density function (pdf) at position x at time t. The L_{FP} operator

$$L_{\mathrm{FP}} = \frac{\partial}{\partial x} \left(\frac{V'(x)}{m\eta_\gamma} + K_r \frac{\partial}{\partial x} \right) \tag{16}$$

with the external potential $V(x)$ [6] contains the anomalous diffusion constant K_γ, and the anomalous friction coefficient η_γ with the dimension $[\eta_\gamma] = s^{\gamma-2}$; herein, m denotes the mass of the diffusion particle. And

$${}_0D_t^{\alpha} W(x,t) = \frac{1}{\Gamma(1-\alpha)} \frac{\partial}{\partial t} \int_0^t \mathrm{d}\tau \frac{W(x,\tau)}{(t-\tau)^{\alpha}}, \qquad 0 < \alpha < 1. \tag{17}$$

Applying Laplace transformation to (15), it becomes

$$s^{\alpha} W(x,s) = G \frac{\partial}{\partial x} \left[\frac{V'(x)}{m\eta_\gamma} + K_\gamma \frac{\partial}{\partial x} \right] W(x,s). \tag{18}$$

Anomalous diffusion in the homogeneous fractal medium in one dimension is characterized by the occurrence of a mean-square displacement of the form

$$\langle (\Delta x)^2(t) \rangle_0 = \frac{2K_\gamma}{\Gamma(1+\gamma)} t^{\gamma} \tag{19}$$

when no external driving force is applied to the particle. The extraction of moments $\langle(\Delta x)^n\rangle$ for anomalous diffusion on fractal medium with dimension d_f is given by [7]

$$\langle(\Delta x)^n(t)\rangle = \int_0^\infty \mathrm{d}x \cdot x^{d_f-1} x^n W(x,t). \tag{20}$$

When the particle is assumed in a constant force field, say $V(x) = -Fx$, by the moment expression (equation (20)) for order one and from the solution of (18), we have the displacement of the particle

$$\langle(\Delta x)(t)\rangle_F = \frac{F\Gamma(d_f+1)}{m\eta_\gamma\Gamma(d_f+2)\Gamma(1+\gamma)} t^\gamma. \tag{21}$$

Thus, from (19), (21) and the generalized Enistein–Stocks–Smoluchowski relation [5]

$$K_\gamma = \frac{k_B T}{m\eta_\gamma}, \tag{22}$$

we get the GER

$$\langle(\Delta x)(t)\rangle_F = \frac{1}{d_f+1}\frac{F\langle(\Delta x)^2(t)\rangle_0}{k_B T}. \tag{23}$$

It shows that the GER holds for FFPE (equation (15)), and that the pre-factor of GER is not a universal constant, while it is given by $d_f + 1$, where d_f is the fractal dimension of the fractal structure considered and $1 < d_f < 2$. This just shows that the numerical pre-factors of the power law should be modified by the local geometry as Amblard *et al* pointed out. In particular, when $d_f \to 1$, it reduces to

$$\langle(\Delta x)(t)\rangle_F = \frac{1}{2}\frac{F\langle(\Delta x)^2(t)\rangle_0}{k_B T}. \tag{24}$$

This is just the result of [5].

4. Conclusion

It is proved that the theory presented by Amblard *et al* is not consistent with the GER. The theory in terms of a fractional Fokker–Planck equation (FFPE) can be well modelled for the experiment by Amblard *et al*. The result obtained by FFPE is consistent with the statement by Amblard *et al* [1] that the numerical pre-factors of the power law would be modified by local geometry and are not exact.

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