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Intermittent Brownian Dynamics over Strands

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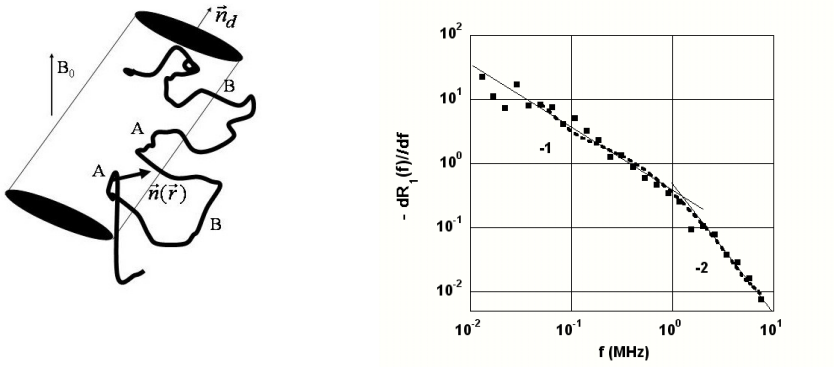
1. Introduction

Intermittent dynamics is intrinsically involved for particles or molecules exploring confining interfacial systems such as porous material, colloidal suspension and catalytic surfaces. Periods of relocation in the bulk phase or in the pore network (bridges) alternate with adsorption or trapping steps generally located nearby the interface (cf. Fig 1A). A representative example concerns the search for a specific target site on DNA by a protein, which alternates adsorption or scanning phases where the protein diffuses on the DNA strand and three-dimensional bulk excursions or relocations [1]. A better understanding of such dynamics is needed in order, for example, to optimize the intermittent search strategy [2]. Recently [3], we have proposed a theoretical analysis of field cycling NMR dispersion technique (NMRD) experiments allowing to probe the time/frequency dependence of relocation steps [4] and adsorption periods of an intermittent fluid dynamics near an interface. In this communication, we consider the Brownian dynamics of a fluid molecule over thin and very long mineral strands having a diameter of about 3 nm. Experiments are compared with numerical simulations and theoretical derivation.

2. Probing relocation statistics over strands by NMR relaxometry



A **B**

Fig.1: **A**: Intermittent Brownian dynamics over a strand. A and B stand for the Adsorption step and the Relocation (Brigde statistics). **B**: Frequency dependence of the spin-lattice relaxation rate in diluted suspension of imogolite. Full squares: experimental results. Dotted line: Brownian dynamics numerical simulation.

In the following, we consider the case of imogolite particles in very diluted suspensions. Imogolite are thin and very long cylinders having a diameter similar to DNA

molecules. Using NMRD, we observe characteristic dispersion curves, $R_1(\omega)$, vastly different from that found for flat or curved interface[5]. As shown in Fig 1B, $R_1(\omega)$ evolves $-\ln(\omega)$ at low frequency. A crossover to a $1/\omega$ is observed above 1 MHz. Brownian dynamics simulation is found to be in good agreement with experimental results, as show in Fig 1B.

3. Theoretical analysis.

We have shown [3] that an intermittent Brownian dynamics induces a low frequency dependence of $R_1(\omega) \propto J(\omega)+4J(2\omega)$ with:

$$J(\omega) = \frac{1}{\omega^2}\mathrm{Re}al\left(\frac{(1-\tilde{\psi}_R(\omega))(1-\tilde{\psi}_A(\omega))}{1-\tilde{\psi}_R(\omega)\tilde{\psi}_A(\omega)}\right) \qquad \text{Eq(1)}$$

$\tilde{\psi}_R(\omega)$ and $\tilde{\psi}_A(\omega)$ are the Fourier transforms of the probability density function characterizing the relocation and the adsorption step, respectively. Using classical properties of the potential theory in 2D, it is possible to show that for a cylinder, at long time, we have $\psi_R(t) \propto 1/t(\ln^2(t))$. The $1/t$ evolution determines a $-\ln(\omega)$ evolution of $J(\omega)$ at small frequency. At short time, the diffusor can only probe the local "flat" surface of the cylinder and then evolves as $1/t^{3/2}$ [4]. This last property induces an algebraic decay of $J(\omega)$ at large frequency [3,5]. These two theoretical results explain in a closed form the experimental data.

4. Conclusion

We have investigated the slow fluid molecule dynamics near strands by field cycling NMR relaxometry. It is a way to follow dynamical correlation from 1 ns to 10 μs. We have shown that the Brownian relocation step is specific of the strand shape. Implication in understanding intermittence Brownian dynamics over DNA (where an unexpected $-\ln(\omega)$ is also observed [6]) is undertaken. Influence of particle concentration and particle conformation are also currently investigated.

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References

[1] M. Coppey, O. Benichou, R. Voituriez, M. Moreau, Biophysical Journal, 87, 1640 (2004)
[2] M. Shlesinger, Nature 443, 281 (2006)
[3] P. Levitz, J. Phys. Condens. Matt. 17, S4059 (2005)
[4] P. Levitz, D. S. Grebenkov, M. Zinsmeister, K. Kolwankar, B. Sapoval, Phys. Rev. Lett. 96, 180601 (2006).
[5] P. E. Levitz, J-P Korb, Europhysics Letters 70, 684.(2005)
[6] V. K.G., Teng C.L., Diensen T.R.D, Korb J-P and Bryant R.G., Magnetic Resonance in Chemistry, 42: 518-523, (2004)